Abstract Booklet

(Last name order, A-Z)

Many thanks to our plenary, invited and contributing speakers and poster presenters

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Optical control of 4*f* orbital state in rare-earth metals

Nele Thielemann-Kuehn[1], Tim Amrhein[1], Wibke Bronsch[1], Somnath Jana[2], Niko Pontius[2], Robin Y. Engel[3], Piter S. Miedema[3], Dominik Legut[4], Karel Carva[5], Unai Atxitia[1], Benjamin E. van Kuiken[6], Martin Teichmann[6], Robert E. Carley[6], Laurent Mercadier[6], Alexander Yaroslavtsev[7,8], Giuseppe Mercurio[6], Loic Le Guyader[6], Naman Agarwal[6], Rafael Gort[6], Andreas Scherz[6], Siarhei Dziarzhytski[3], Guenter Brenner[3], Federico Pressacco[3], Ru-Pan Wang[3], Jan Schunck[3], Mangalika Sinha[3], Martin Beye[3], Gheorghe Sorin Chiuzbaian[9], Peter M. Oppeneer[8], Martin Weinelt[1], Christian Schueßler-Langeheine[2]

[1]Freie Universität Berlin, Berlin, Germany. [2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany. [3]Deutsches Elektron-Synchrotron DESY, Hamburg, Germany. [4]IT4Inovations-Czech National Supercomputing Centre, VSB - Technical University Ostrava, Ostrava, Czech Republic. [5]Charles University, Faculty of methematics and Physics, DCMP, Prague, Czech Republic. [6]European XFEL, Schenefeld, Germany. [7]European XFEL, Berlin, Germany. [8]Uppsala University, Department of Physics and Astronomy, Uppsala, Sweden. [9]Sorbonne Université, Laboratoire de Chimie Physique Matière et Rayonnement, Paris, France

Abstract

In recent years it has been appreciated that early electronic excitations may play a more important role for ultrafast spin physics than just being a transient step in the deposition of energy in the electron system [1,2]. However, in the case of 4*f* metals, optically induced *f*-*f* electronic excitations within the 4*f* shell are dipole

forbidden.

Recently, a time-resolved X-ray absorption spectroscopy experiment at European XFEL gave us an indication on a $5d$-$4f$ electron-electron scattering process leading to excited $4f$ states in Tb metal on ultrafast time scales [3].

To further investigate these excited $4f$ states we performed a time-resolved resonant inelastic X-ray scattering experiment at the free electron laser FLASH I. We present our work on Tb and show the fingerprints of these ultrafast $4f$ excitations.

Altering the electronic state and hence the spin and orbital momenta in the $4f$ electronic system changes its coupling to the environment. This provides a new handle to transiently alter material properties like the magneto crystalline anisotropy.

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Indication of a quantum critical point in cuprate high-Tc superconductors via Resonant Inelastic X-ray Scattering

Riccardo Arpaia[1], Leonardo Martinelli[2], Marco Moretti Sala[2], Sergio Caprara[3], Abhishek Nag[4], Nicholas B. Brookes[5], Pietro Camisa[2], Qizhi Li[6], Ying Ying Peng[6], Mirian Garcia-Fernandez[4], Kejin Zhou[4], Thilo Bauch[1], Carlo Di Castro[3], Marco Grilli[3], Floriana Lombardi[1], Lucio Braicovich[2], Giacomo Ghiringhelli[2]
[1]Chalmers University of Technology, Göteborg, Sweden. [2]Politecnico di Milano, Milano, Italy. [3]Università "La Sapienza", Roma, Italy. [4]Diamond Light Source, Didcot, United Kingdom. [5]The European Synchrotron, Grenoble, France. [6]Peking University, Beijing, China

Abstract

The strange metal phase, which dominates large part of the phase diagram of the cuprates [1], exhibits a certain degree of universality since it has been found in many different materials, including pnictides, heavy fermions and magic angle double layer graphene. One suggested origin of this universality is that it stems from a quantum critical point (QCP), i.e., a phase transition at zero temperature driven by quantum fluctuations, in contrast to ordinary phase transitions driven by thermal fluctuations [2]. However, the evidence for the existence of a quantum critical point (QCP) in the phase diagram of cuprates remains elusive. This is mostly due to the challenge to identify the nature of the quantum fluctuations which drive the novel physics of the strange metal phase.

Here we show that the recently discovered charge density fluctuations (CDF) [3,4] have several properties required to be associated to a quantum phase transition. By using resonant inelastic x-ray scattering (RIXS), we have determined the intensity and the energy of the CDF in two families of cuprate high-Tc

superconductors, at doping levels, ranging from the strongly underdoped regime up to the putative QCP at p* ≈ 0.19 [5]. The determination of the characteristic energy is a challenge, as it is of the order of few tens of meV, i.e. CDF largely overlap with phonons and other charge excitations. Therefore very high resolution is needed, together with sample position stability over a wide temperature range.

We show that the CDF intensity is strongest at p*, while it softens at lower dopings. More importantly, its characteristic energy increases when moving away from p*, following a wedge shape typical of a quantum critical region and in agreement with what observed in the cuprate phase diagram. These results support the leading role of charge order in driving the anomalous properties of the strange metal, adding new clues to the mechanism for high critical temperature superconductivity [6].

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Novel Multi Atom Resonance X-ray Raman Scattering - from theory and experimental approaches

Kiyotaka Asakura[1], Kaori Niki[2], Takashi Fujikawa[2]

[1]Institute for Catalysis, Hokkaido Universit, Sapporo, Japan.

[2]Graduate School of Advanced Integration of Science, Chiba University, Chiba, Japan

Abstract

Consider a Pt nanoparticle catalyst and the investigation of its active structure under the reaction conditions, where CO or oxygen might be adsorbed on its surface. Although Pt L_3 edge operando X-ray absorption fine structure (XAFS) provides Pt local structures, CO or oxygen XAFS measurements under the reaction conditions are difficult for two reasons; One is too strong absorption of gas phase and the other is the overlapping of the gas phase XAFS.

If we tuned X-ray energy to the Pt L_3 edge and the absorbed energy is transferred to CO or O- adsorbates to excite them to produce the Raman emission, we could observe the CO or O XAFS signal at high energy with less interference from gas phase. The method is named as multi atom resonance X-ray Raman (MARX-Raman) XAFS for the analogy of MARPE(multi atom resonance photoemission)[1].

We found the evidence of MARX-Raman in $Eu(CP)_3$ (CP=cyclopentadiene).[2] When the X-ray incidence(excitation) energy was tuned to Eu L_1-edge at 9752 eV and the scattering X-ray was detected around the energies 9480-9450 eV, corresponding to the loss energy of C K-edge, we found the abrupt increase in the emission intensity at about 9467 eV or loss energy of 285 eV, indicating the possibility for MARX-Raman.

We theoretically studied MARX Raman and obtaibed

following formula.[3]

$$I(\lambda',\lambda) \approx -\frac{2}{(E_0+\omega_\lambda-E_0^*)^2+\Gamma^2/4}\sum_{rm}\beta_{rr}|\Delta_{rd}(\lambda)|^2$$
$$\times|R_m|^2|<\phi_d|t_{1m}|\phi_r>|^2$$
$$\times\mathrm{Im}\Big(<c|\Delta^\dagger(\lambda')g_c(E_0+\omega_\lambda-\omega_{\lambda'}-E_0^{**})\Delta(\lambda')|c>\Big) \quad (1)$$

The formula is quite similar to the MARPE.[4] MARX-Raman and MARPE cannot be observed in a high symmetric environment owing to (1). The difference is the dependence of bond distance. In MARPE R_m is proptional to $1/r^2$ while R_m is proptional to $1/r^3$ in MARX Raman as shown in (1).

Although we have indicated the possibility of MARX-Raman theoretically and experimentally, we need further experimental evidence to demonstrate the availability as a new operando XAFS technique.

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Direct measurement of 3d-4s excitations in a linearly coordinated Fe1+ single-ion magnet

Myron Huzan, Michael Baker
The University of Manchester, Manchester, United Kingdom

Abstract

Linearly coordinated transition metal sites are predicted to exhibit non-intuitive ligand-field splitting due to strong $3d_{z2}$-4s hybridisation. Conventional ligand-field theory predicts the energy of the $3d_{z2}$ orbital to increase due to a pair of bonds acting along the z-axis. The $D_{\infty h}$ ligand field splitting for this scenario is shown in figure 1, where the $3d_{z2}$ orbital has higher energy than the $3d_{xz,yz}$ and the $3d_{x2y2,xy}$ orbitals have the lowest energy. However, in practice, the ligand field is strongly perturbed due to $3d_{z2}$-$4s$ hybridisation. The result is a lowering in the energy of the $3d_{z2}$ orbital and a significant amount of $3d_{z2}$ orbital character being mixed into the $4s$ shell. I will present why direct observation of $3d_{z2}$-$4s$ hybridisation is challenging to access by X-ray absorption and that it is only via high-resolution $2p3d$ RIXS measurements, conducted at I21 at Diamond Light Source, that access to $3d_{z2}$ mixing enhanced $4s$ excitations can be observed in Fe doped lithium nitride (structure shown in figure 1)[1]. The $3d_{z2}$-$4s$ hybridisation strongly implicates the magnetism of linearly coordinated transition metal ions and in the presented case of iron-doped lithium nitride is crucial to the observed single-ion magnet behaviour present.

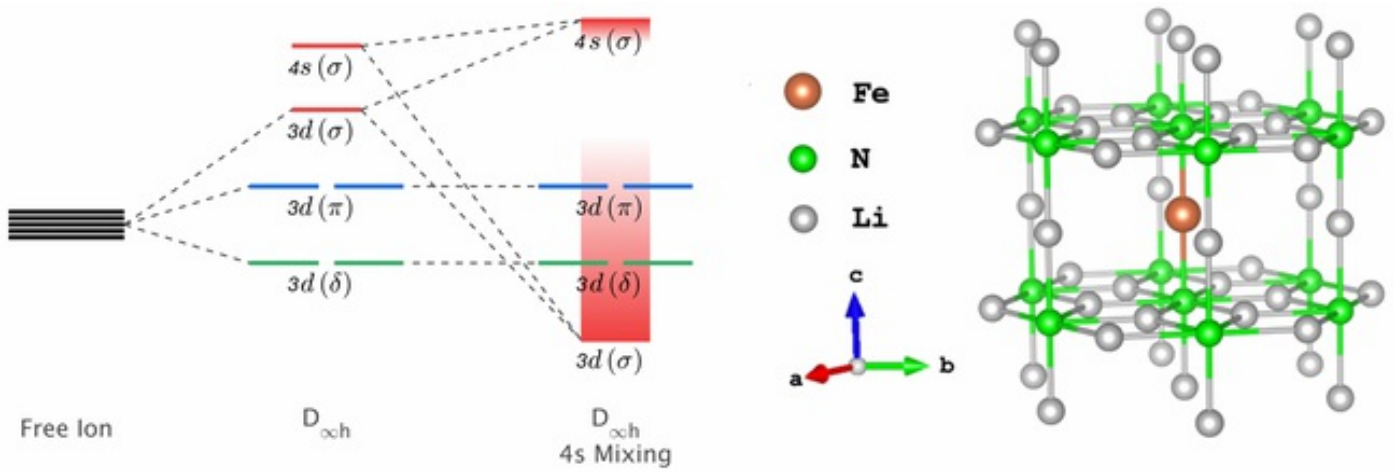

Figure 1. Left: Schematic showing *3d* orbital energy non-degeneracy due to a linear ligand field and the influence of *3d-4s* mixing. Right: The structure of iron-doped lithium nitride, where Fe dopant sites replace Li-ions with a linear coordination environment[2].

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Identifying Redox Orbitals in Lithium-Ion Cathodes with x-ray Compton Scattering

Bernardo Barbiellini
LUT University, Lappeenranta, Finland

Abstract

The reduction-oxidation (redox) reactions are fundamental processes taking place in Li-ion batteries. Thus, the study of the redox orbitals provides insight at the atomic scale into the cathode working. The research reported in this presentation involves x-ray Compton Scattering, and computational methods based on Density Functional Theory capable of identifying redox orbitals. The main goal is to extract the electronic states that lead to better performances in Li-ion batteries [1-4].

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Hydrodynamic Interaction Between Quasi-elastic and Acoustic Modes Observed by Inelastic X-Ray Scattering

Alfred BARON[1,2], Daisuke ISHIKAWA[3,2]

[1]MateriaMaterials Dynamics Laboratory, RIKEN SPring-8 Center, Sayo, Japan. [2]Precision Spectroscopy Division, CSRR, SPring-8/JASRI, Sayo, Japan. [3]MateriMaterials Dynamics Laboratory, RIKEN SPring-8 Center, Sayo, Japan

Abstract

Improvements in methodology and instrumentation for meV-resolved inelastic x-ray scattering (IXS) [1–4], coupled with a fresh examination of older theory (e.g. [5]), allow identification of an interaction between the quasi-elastic and acoustic phonon modes in liquids [6]. This helps explain a decades old controversy about the appearance of additional modes in water spectra, and provides a strong base to discuss new phenomena in liquids on the mesoscale. Further, we find that the intensity of the quasi-elastic peak in water is in good quantitative agreement with an estimate from a simple viscoelastic model [7] using the magnitude of the positive dispersion, so the huge increase in the quasi-elastic scattering in water on the mesoscale (relative to the long wavelength Landau-Placzek ratio) can robustly be interpreted as being due to the relaxation that causes fast sound [6]. We also show that including the interaction changes the fit results, resulting in significant changes to the observed sound velocity relative to a model without interaction.

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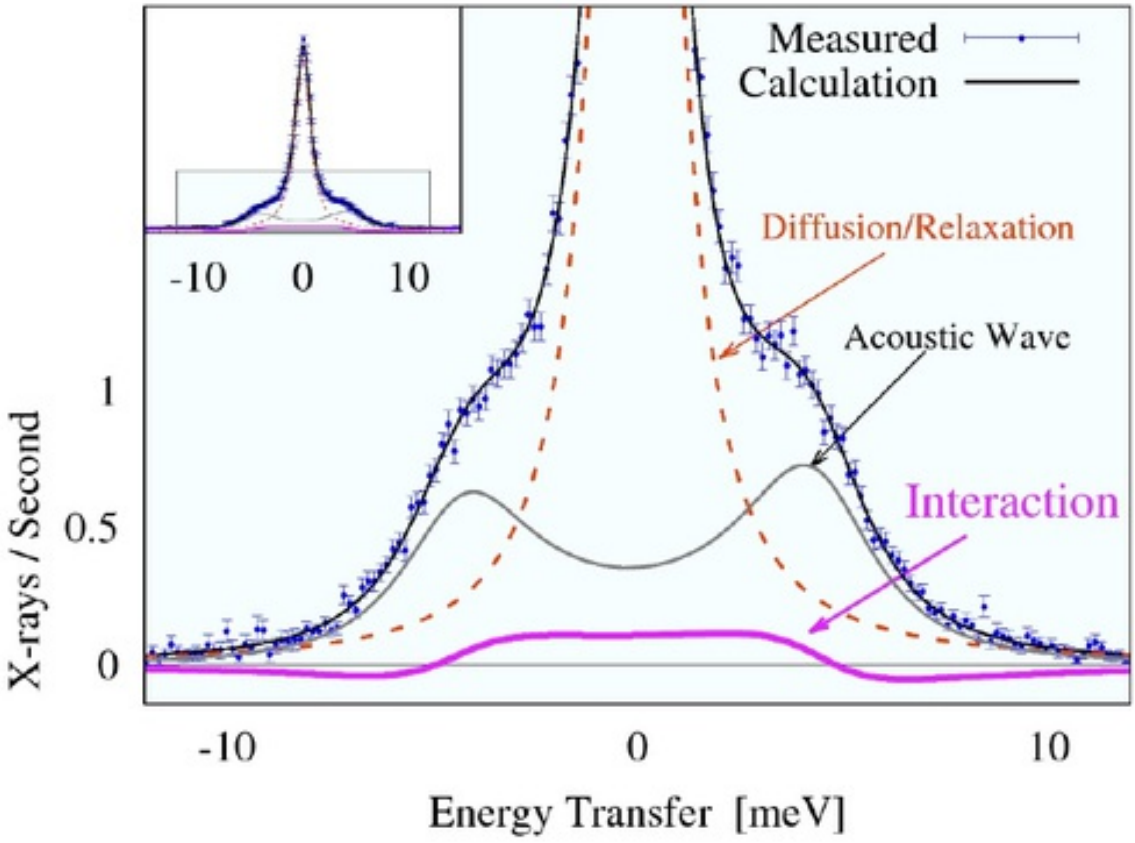

IXS from water at 301K at Q=2.5 nm^{-1}. The various contributions are as indicated. A fit with the full interacting model (black line) gives small residuals (reduced chi-squared 0.92, probability>0.5) while a fit without interaction gives large and correlated residuals (reduced chi-squared 1.56, probability<10^{-5}). [6]

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Inelastic X-ray scattering to study Cultural and Natural Heritage Samples

Loïc Bertrand[1], Rafaella Georgiou[1], Serge Cohen[1], Lauren Dalecky[1], Laure Cazals[1], Sylvain Bernard[2], Pierre Gueriau[1], Dimosthenis Sokaras[3], Alessandro Mirone[4], Christoph Sahle[4], Jean-Pascal Rueff[5], Uwe Bergmann[6]
[1]Université Paris-Saclay, Gif-sur-Yvette, France. [2]Muséum national d'histoire naturelle, Paris, France. [3]SLAC, Stanford, USA. [4]ESRF, Grenoble, France. [5]Synchrotron SOLEIL, Gif-sur-Yvette, France. [6]University of Wisconsin, Madison, USA

Abstract

The analysis of materials from art, archaeology, palaeontology and paleoenvironmental research requires methods capable of dealing with heterogenous, altered and rare systems [1,2]. Among them, ancient organic compounds are very delicate to decipher, especially if one wants to avoid sampling or cleaning while limiting the effects of surface alteration and contamination. An ideal method would identify the speciation of organic compounds in 3D in a bulk sample. Here, we describe recent results obtained by exploring the organic chemistry of several ancient samples. X-ray Raman spectroscopy (XRS) has allowed us to study the organic speciation of the skin of a 49,000-yr-old mammoth from Siberia [3], of a 56-Myr-old fossil cocoon [3], and of a set of plant exudates (resins, kinos and gums) from a historical collection documenting Aboriginal Australians uses [4]. XRS raster scanning imaging has provided insight in the preservation pathway of a 305-Myr-old wood sample [5]. In addition, we have shown, with the study of the preservation of a 53-Myr-old ant trapped in amber [5], that direct tomography developed by Huotari et al. [6] could be transposed to ancient systems. While a number of synchrotron instruments could be used to study complex heritage and/or natural samples [7], the main limitation of the

approach is the low cross-section of inelastic X-ray scattering, which requires mitigating radiation-induced side effects [8].

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Compton scattering as a possible probe to reveal hidden real-space and reciprocal-space magnetoelectric multipoles

Sayantika Bhowal[1], Stephen Collins[2], Nicola Spaldin[1]
[1]ETH Zurich, Zurich, Switzerland. [2]Diamond Light Source Ltd, Didcot, United Kingdom

Abstract

The magnetoelectric multipoles quantify the antisymmetric magnetization density of a solid that results from broken space-inversion I and time-reversal τ symmetries, and play a crucial role in the understanding of the linear magnetoelectric response in a solid, describing the linear order magnetization induced by an applied electric field and vice-versa. In my talk, using symmetry analysis and density functional calculations, I will discuss the possibility of applying (designing) regular and magnetic Compton scattering experiments to detect the real-space and reciprocal-space magnetoelectric multipoles, which often escape detection in conventional probes. Compton scattering of x-ray photons was an early confirmation of quantum mechanical behavior in its revolutionary step of assigning momentum to electromagnetic waves. The effect is widely used today in fields as diverse as radio-biology, astrophysics, and condensed matter, where it is used to measure the distribution of electron density in momentum space. While the extension to measuring the spin-dependent momentum distribution, now known as magnetic Compton scattering, was proposed as early as 1970, dramatic advances in synchrotron light sources have now made it possible to capture the weak scattering cross-section in the spin channel of different materials. We show that due to the non-trivial duality between real space and momentum space, the antisymmetric magnetization density of a real-space magnetoelectric multipole transforms into an antisymmetric electron density in momentum space, resulting in an anti-symmetric signal in the regular Compton profile. As a counterpart of the real space-

momentum space duality, the antisymmetric electron density in a broken inversion symmetric system (quantified by an odd-parity electric multipole) transforms into an antisymmetric magnetization density in reciprocal space. Such antisymmetric spin momentum density is representative of the magnetoelectric multipoles, although in k-space, and consequently gives rise to an antisymmetric magnetic Compton profile in an apparently nonmagnetic system. Taking the examples of the well-known magnetoelectric material LiNiPO4 [1] and the prototype ferroelectric PbTiO3 [2], I will discuss the role of various material properties that control the magnitude and direction of the antisymmetric Compton profiles.

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Acknowledgements:

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Functional quantum materials studied with resonant inelastic x-ray scattering

Valentina Bisogni

Brookhaven National Laboratory
bisogni@bnl.gov

The past years have witnessed an increasing interest in the field of quantum materials (QM), not only thanks to their fascinating behaviour as a macroscopic manifestation of quantum mechanics, but also for the opportunities that these materials offer in terms of functional properties (high-temperature superconductivity, quantum Hall effect, giant magnetoelectric effect, *etc...*). New types of electronics can be envisaged using QMs. Magnonics is one of them, where the transport is mediated by collective spin excitations (magnons, spinons, *etc...*), yielding low-power and energy-efficient electronics.

Studying spin excitations and their dynamics in functional quantum materials, either in the ground state form or under device-operating conditions, is thus an important step from both a fundamental and an applied perspective. In this talk, I will present recent results on spin excitations in magnonic model-devices using soft Resonant Inelastic X-ray Scattering. More specifically, I will focus on: *i)* the evolution of the spin dynamics as a function of thickness in magnetic thin films [1]; *ii)* the visualization of the magnon mode contributing to the magnon current in Yttrium Iron Garnet, with energy- and momentum-resolution [2].

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Probing lattice oxygen oxidation in $SrFeO_3$ during oxygen evolution by RIXS

Jan Bosse
Paul Scherrer Institute, Villigen, Switzerland

Abstract

Perovskite oxides (ABO_3) have emerged as a promising class of electrocatalytic materials for water oxidation due to their easily tunable electronic structure. However, under electrochemical non-equilibrium conditions of water oxidation, transition-metal perovskites can intercalate oxygen and increase the transition metal oxidation state. This process can potentially result in the **lattice oxygen oxidation**, which can cause amorphization due to the collapse of the crystal lattice.

I will show how these fundamental processes are linked to the electrochemical response in the pre-OER and OER potential region of a representative perovskite $SrFeO_3$. Using epitaxial thin films of $SrFeO_{3-x}$ as a model system, I will discuss the chemical and structural changes in the potential range up to the OER. $SrFeO_{3-x}$ experiences a brownmillerite-to-perovskite transition during oxygen intercalation ($SrFeO_{2.5}$ into $SrFeO_3$, fig. 1), which can lead to oxidation states of the transition metal that cannot be accommodated by the electronic structure without the oxidation of lattice oxygen. I will present RIXS results that will address the possibility of lattice oxygen oxidation and its role in the degradation of the perovskite structure during OER.

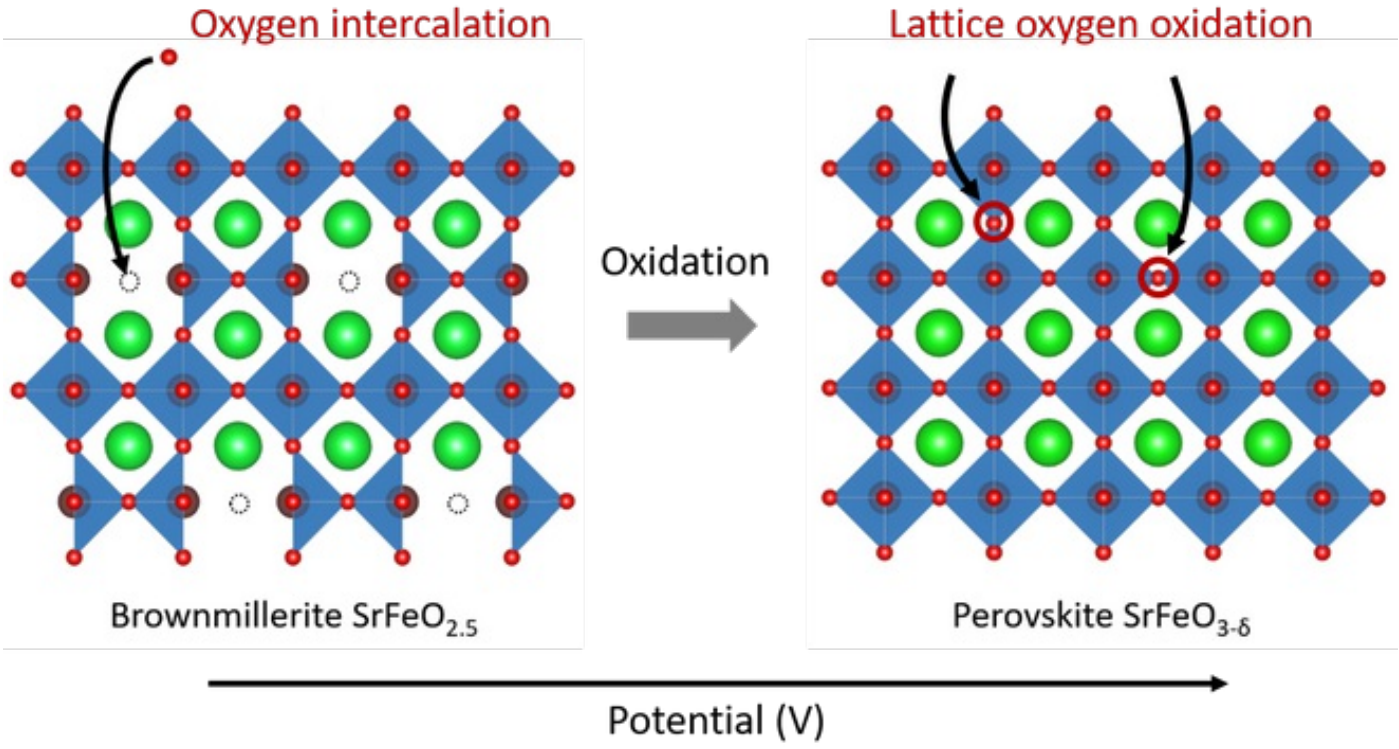

Fig. 1: The transition from brownmillerite $SrFeO_{2.5}$ to perovskite $SrFeO_3$ during oxygen intercalation.

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Spin Waves in FCC-cobalt

Fillipo Elgorni[1], Davide Betto[1], Roberto Sant[1], Kun Cao[2], Nicholas Brookes[1]
[1]ESRF, Grenoble, France. [2]Sun Yat-sen University, Sun Yat-sen, China

Abstract

Spin fluctuations in magnetic materials have been studied over many decades but are still of relevance today, for example in current research areas of superconductivity, magnonics and spintronics. Theory has advanced greatly in describing such phenomena but still needs to improve [1,2]. Studying simple ferromagnetic metals like iron, cobalt, nickel with inelastic neutron scattering allowed spin wave dispersion to be observed and the Landau damping resulting from the interaction with the Stoner continuum, to be studied. This has been a testing ground for theory and the methods are still improving with modern state of the art calculations, which can include the spin fluctuations [2].

Recently it has been shown that soft x-ray RIXS can be used to study spin waves in metallic ferromagnets like iron and nickel [3, 4]. This has been achieved with nanometre thick thin films and has even allowed the study as the number of atomic layers have been reduced [4]. These studies expand the range of problems that can be addressed by soft x-ray RIXS.

Here we will discuss the application of soft x-ray RIXS to thin fcc-cobalt films and compare the spin-wave dispersion to TD-DFPT calculations as in Ref. [2].

The calculations have been performed as described in Ref. [3] using the time-dependent density functional perturbation theory

implemented on top of Quantum ESPRESSO [5]. This comparison between experimental data and theory allows us to gain a better understanding of the behaviour of spin waves in purely metallic systems, in view of future magnonics and spintronics applications.

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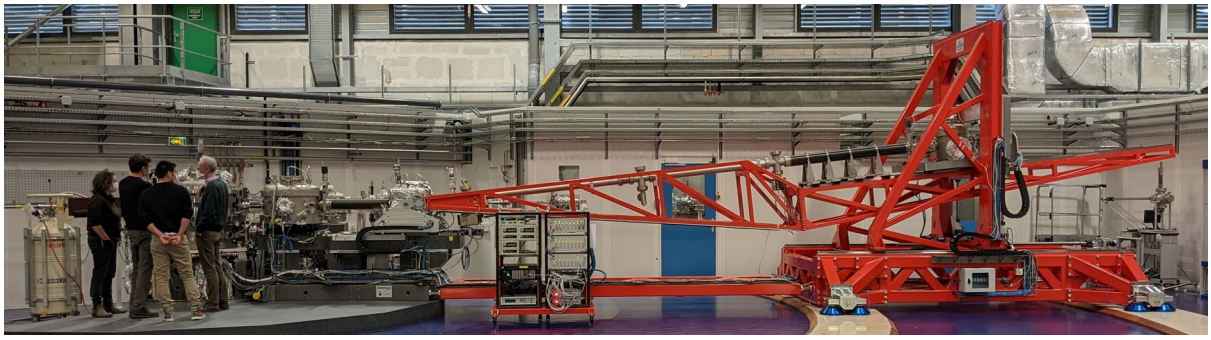
The resolution upgrade of the ESRF RIXS spectrometer at ID32

K. Kummer,[1] R. Barrett,[1] D. Betto,[1] C. Clavel,[1] L. Eybert-Berard,[1] G. Ghiringhelli,[2,3] L. Martinelli,[2] R. Sant,[1] A. Vivo,[1] F. Yakhou-Harris,[1] N.B. Brookes[1]

[1] *ESRF*
[2] *Dipartimento di Fisica, Politecnico di Milano, piazza Leonardo da Vinci 32, I-20133 Milano, Italy*
[3] *CNR-SPIN, Dipartimento di Fisica, Politecnico di Milano, I-20133 Milano, Italy*

The soft X-ray RIXS spectrometer at the ID32 beamline underwent an upgrade during the EBS shutdown. As a result the total energy resolution of the instrument improved by about 25% over the entire energy range from 400 to 1600 eV. The previously available energy resolution can now achieved with much more efficient optics improving the through-put by a factor 3 to 4 compared to the pre-EBS instrument. At the Cu L_3 edge, an energy resolution as high as 25 meV and polarisation resolved RIXS measurements with an energy resolution as good as 40 meV are now available[1]. The upgraded spectrometer has been in user operation since June 2020.



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Email corresponding author: brookes@esrf.fr

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Revealing mechanisms of charge storage in Li and Na battery materials with RIXS and other techniques

Peter Bruce
Universtiy of Oxford, Oxford, United Kingdom. Henry Royce Institute, Oxford, United Kingdom. Faraday Institution, Harwell, United Kingdom

Abstract

Revealing mechanisms of charge storage in Li and Na battery materials with RIXS and other techniques

Peter G Bruce1,2,3

1Departments of Materials and Chemistry, University of Oxford

2The Henry Royce Institute, Parks Road, Oxford, OX1 3PH, U.K.

3The Faraday Institution, Quad One, Becquerel Avenue, Harwell Campus, Didcot, OX11 0RA, U.K.

Energy materials research is a critical part of the global effort to tackle the climate crisis. In particular, advances in battery technology hold the key to electrification of our transport systems and to decarbonisation of the grid. In the endeavour to achieve new energy materials with breakthrough performance, we must first understand the fundamental science which underpins how these materials function.

In this talk, I will outline how advanced materials characterisation tools, especially RIXS, have revealed the origin of exceptional levels of charge storage in Li-ion battery cathodes. I shall show how RIXS data enabled us to identify the formation of O2 molecules trapped within the cathode particles as a result of O2- oxidation, explaining

charge storage beyond the limits of conventional cathodes and offering an explanation of the voltage hysteresis observed in this class of materials.1

The RIXS results were complemented by other techniques, such as SQUID2 and neutron PDF3, and they have led to a general model to explain the oxygen redox reaction.4 Furthermore, it is possible to suppress O2 formation, trapping hole states on O2- and obtaining energetic (voltage) and structural reversibility.5 Such behaviour points the way towards high energy density cathodes for Li-ion batteries.

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The meV-resolved Inelastic X-ray Scattering Spectrometer at NSLS-II: Recent Performance Improvement and Science Capabilities Update

Yong Cai[1], Alexey Suvorov[1], Scott Coburn[1], Kazimierz Gofron[1], Alessandro Cunsolo[1,2]

[1]NSLS-II, Brookhaven National Laboratory, Upton, NY 11973, USA.

[2]Department of Physics, University of Wisconsin-Madison, Madison, WI 53706, USA

Abstract

The ultrahigh resolution inelastic x-ray scattering (IXS) beamline at NSLS-II is designed to achieve ~meV resolution at a moderate energy of ~9 keV for IXS experiments with high momentum resolution and high spectral contrast [1]. The key instrument is a novel spectrometer with a new type of analyzer optics based on post-sample collimation coupled with angular dispersive flat crystal optics in highly asymmetric Bragg reflections [2,3] to achieve comparable angular acceptance of the scattered photons as that of the spherical backscattering analyzer used in other meV-resolved IXS facilities operating at above 20 keV. As the first user instrument of its kind, the spectrometer has demonstrated unique research capabilities in studying low-Q, THz dynamics in material systems with mesoscopic heterogeneity and complexity including most soft materials and biological systems [4,5]. The relatively low operation energy furthermore offers enhanced sensitivity in studying systems of reduced dimensionalities including surfaces of single crystals and thin films as shown in recent experiments in quantum topological materials. The recent 10X improvement in data rate at an energy resolution of less than 1.9 meV has enabled the latter class of experiments to be performed routinely at the beamline. In this presentation, details on the operation and performance of the spectrometer as well as potential future upgrades to enhance substantially the data efficiency further will be presented and

discussed.

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† E-mail: cai@bnl.gov

‡ Current address: Department of Physics, University of Wisconsin-Madison, Madison, WI 53706, USA

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Revealing the nature of stripe order in cuprate superconductors.

Joahn Chang
University of Zurich, Zurich, Switzerland

Abstract

This talk will present our most recent RIXS experiments designed to uncover the nature of stripe order in La-based cuprate superconductors [1-4].

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Operando 2p3d RIXS of $La(Ni,Fe)O_3$ ultrathin films during electrocatalysis

Qijun Che[1], Masoud Lazemi[1], Emma van der Minne[2], Iris van den Bosch[2], Yorick A. Birkhölzer[2], Moritz Nunnenkamp[2], Matt Peerlings[1], Christoph Baeumer[2], Gertjan Koster[2], Frank de Groot*[1]
[1]Materials Chemistry and Catalysis, Debye Institute for Nanomaterials Science, Utrecht University, Utrecht, Netherlands.
[2]MESA+ Institute for Nanotechnology, University of Twente, Enschede, Netherlands

Abstract

The understanding of the solid-liquid interface is crucial to design rational electrocatalysts for oxygen evolution reactions (OER).[1] Nevertheless, the electricity-driven anodic water splitting makes it complex to identify any dynamical ultrathin surface transformation.[2] Operando X-ray absorption spectroscopy (XAS) and resonant inelastic X-ray scattering (RIXS) provide a great opportunity to study electronic structures such as (co)valence and charge transfer multiplets.[3-4] The fluorescence yield of 2p XAS and 2p3d RIXS is a bulk-sensitive technique, but surface information can be obtained by using very thin layers. Herein, we prepared epitaxial $La(Ni,Fe)O_3$ films with a few nanometers thicknesses on $La_{0.67}Sr_{0.33}MnO_3$ via pulsed laser deposition, which is supported on monolayer $Ca_2Nb_3O_{10}$ (CNO) nanosheet-buffered Si_3N_4 membranes. We are investigating $La(Ni,Fe)O_3$ thin films under operando conditions in the photon-in-photon-out method through the membranes to track the dynamical electronic structure of active materials (Figure 1). The operando Fe and Ni K-edges XAS of an eight-unit-cell $La(Ni,Fe)O_3$ thin films show a significant spectral change under the bias from 0.177 V to 1.077 V (vs. Ag/AgCl) in 1.0 M KOH solution, implying that the structure of the active catalyst changes during the OER (Figure 1 (b,

c)). This suggests that the operando 2p3d RIXS experiment is feasible and allows the study of dd-excitations under working conditions (Figure 1e), where we will combine high-resolution (20-100 meV) 2p3d RIXS as well as low-resolution (1 eV) 2p3d RIXS.

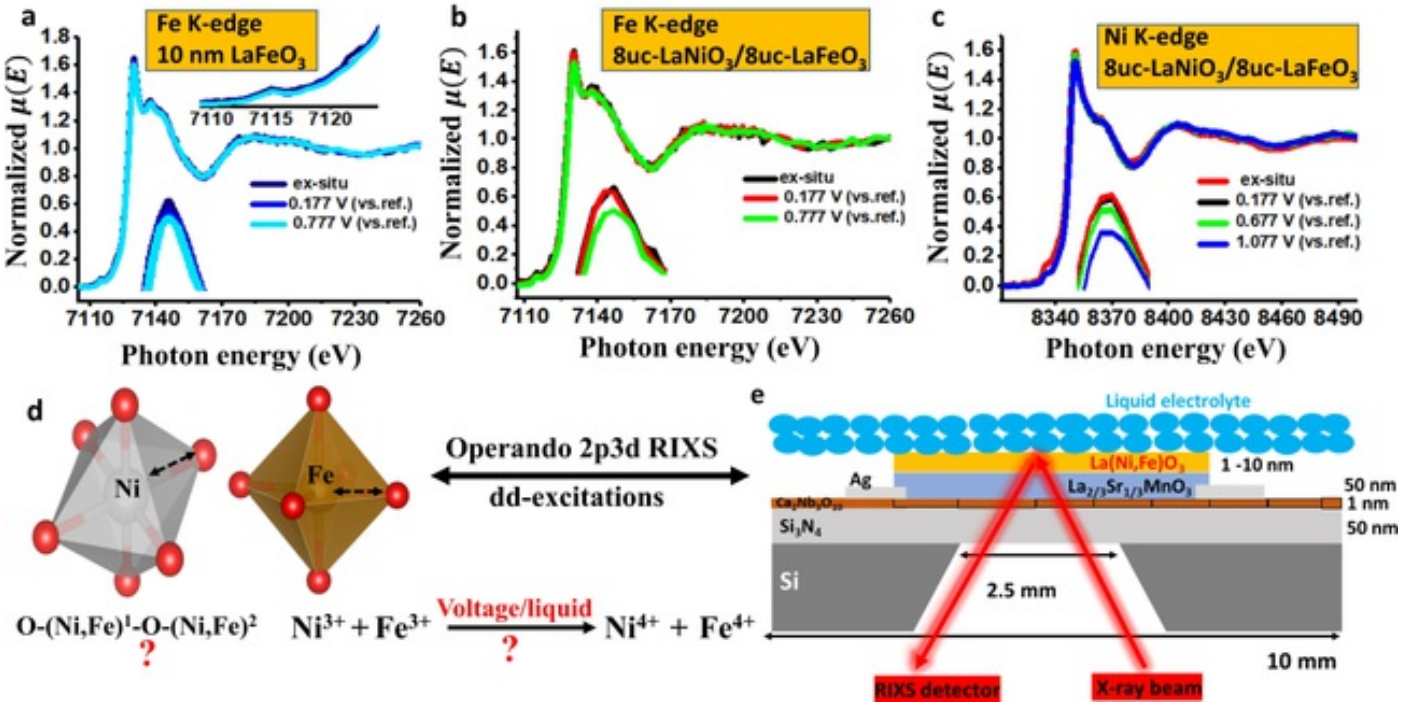

Figure 1. (a-c) Operando Fe and Ni K-edges of X-ray absorption near edge structure (XANES) for La(Ni,Fe)O_3 ultrathin films. (d, e) schematic diagram of operando 2p3d RIXS to track the dynamical electronic structures under the alkaline liquid.

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*Corresponding author: f.m.f.degroot@uu.nl.

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Unveiling Unequivocal Charge Stripe Order in a Prototypical Cuprate Superconductors

Jaewon Choi[1,2], Qisi Wang[1], Simon Jöhr[1], Niels B. Christensen[3], Julia Küspert[1], Damian Bucher[1], Dominik Biscette[1], Mark H. Fischer[1], Markus Hücker[4], Tohru Kurosawa[5], Naoki Momono[6,5], Migaku Oda[5], Oleh Ivashko[7], Martin von Zimmermann[7], Marc Janoschek[8], Johan Chang[1]

[1]Universität Zürich, Zürich, Switzerland. [2]Diamond Light Source, Didcot, United Kingdom. [3]Technical University of Denmark, Lyngby, Denmark. [4]Weizmann Institute of Science, Rehovot, Israel. [5]Hokkaido University, Sapporo, Japan. [6]Muroran Institute of Technology, Muroran, Japan. [7]Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany. [8]Paul Scherrer Institut, Villigen, Switzerland

Abstract

In the cuprates, high-temperature superconductivity, spin- and charge-density-wave (CDW) order are intertwined, and symmetry determination is challenging due to domain formation. We investigated the CDW in the prototypical cuprate $La_{1.88}Sr_{0.12}CuO_4$ via x-ray diffraction employing uniaxial pressure as a domain-selective stimulus to establish the unidirectional nature of the CDW unambiguously. The response of CDW peak along two in-plane directions was markedly different. The intensity of CDW peak perpendicular to the direction of uniaxial pressure application increases with a factor of two, whereas that along the application direction was suppressed completely. This result indicates that the uniaxial pressure removes the degeneracy of unidirectional charge stripe order hosted by two kinds of mutually 90°-rotated domains. A five-fold enhancement of the CDW amplitude is found when homogeneous superconductivity is partially suppressed by a

magnetic field. This field-induced state provides an ideal search environment for a putative pair-density-wave state.

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Elucidating the Symmetry of Charge Density Wave in Cuprate Superconductors via Resonant Inelastic X-ray Scattering

Jaewon Choi[1], Jiemin Li[1,2], Abhishek Nag[1], Pelliciari Jonathan[3,4], Andrew Walters[1], Stefano Agrestini[1], Mirian Garcia-Fernandez[1], Dongjoon Song[5], Hiroshi Aisaki[5], Steve Johnston[6], Riccardo Comin[3], Hong Ding[2], Ke-Jin Zhou[1]

[1]Diamond Light Source, Didcot, United Kingdom. [2]Chinese Academy of Science, Beijing, China. [3]Massachusetts Insititute of Technology, Cambridge, USA. [4]Brooklyn National Laboratory, Upton, USA. [5]National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan. [6]The University of Tennesse, Knoxville, USA

Abstract

Charge density waves (CDW) discovered in copper-oxide superconductors share a universal phenomenology such as their residence in the CuO_2 plane and largest onset temperature at 1/8 hole doping concentration, suggesting a common organizing principle. However, a longstanding debate about whether its spatial symmetry is a unidirectional stripe or a bidirectional checkerboard has hindered the development of a unified picture. Distinguishing these two scenarios is technically difficult, especially in a disordered system with short-range correlation such as Bi-based cuprate superconductors. We employed high-resolution resonant inelastic x-ray scattering on $Bi_2Sr_{1.4}La_{0.6}CuO_{6+\delta}$, a single-layer Bi-based cuprate material, to explore the global and local symmetry of CDW peak structure in the reciprocal space. Our data reveal a globally four-fold diffraction structure, with local CDW peaks elongated along a transverse direction. This result can be best supported by the presence of charge stripe orders, hosted by mutually 90°-rotated anisotropic domains. Our work paves a way for the unified

mechanism underlying CDW phenomenology in this material family.

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Status of the double-dispersion RIXS instrumentation (QERLIN) at the ALS

Yi-De Chuang, Harold Barnard, Alastair MacDowell, Dmitriy Voronov, Valeriy Yashchuk, Daniele Cocco, Azriel Goldschmidt, Peter Denes, Howard Padmore
Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, USA

Abstract

A novel optical scheme for recording resonant inelastic X-ray scattering (RIXS) spectra over a wide excitation photon energy range in one acquisition was proposed. A RIXS instrumentation based on this double-dispersion optical scheme was designed. This scheme requires the beamline monochromator to produce a ~ 2 μm wide by > 2 mm tall stripe of monochromatic X-rays at its focal plane for broad-band excitation. When the sample is placed at this focal plane, different vertical position on the sample will be excited by X-rays with different energy. The spectrometer uses a Wolter mirror pair to re-image the beam profile on the sample vertically onto the detector, and a Hettrick-Underwood dispersive assembly disperses the emitted X-rays from the sample horizontally. The stigmatic focus of the spectrometer ensures the detector to stay in-focus to produce the RIXS map.

The high spectral resolution requirement of this instrumentation pushes the in-house development for grating fabrication and the sCMOS detector with 5 μm square pixels (an array of 2000 X 4000 pixels) for direct X-ray detection. In this talk, I will report the status of this project and challenges we face during the construction.

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Status of the qRIXS instrument at LCLS-II

Georgi Dakovski[1], Frank O'Dowd[1], Jim Defever[1], Margaret Ghaly[1], Joe Dvorak[2]

[1]SLAC National Accelerator Laboratory, Menlo Park, USA.

[2]Brookhaven National LAboratory, Upton, USA

Abstract

The superconducting upgrade of the accelerator structure at LCLS-II promises to deliver 3-to-4 orders of magnitude increase in average flux. This will open the door to comprehensive, high time- and energy-resolution RIXS experiments at our free electron laser. I will present a summary of our efforts in the last several years in designing and constructing qRIXS, a dedicated instrument for studying correlated materials at LCLS-II.

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Probing electron-phonon interactions away from the Fermi level with resonant inelastic x-ray scattering

C. D. Dashwood[1], A. Geondzhian[2,3], J. G. Vale[1], A. C. Pakpour-Tabrizi[1], C. A. Howard[1], Q. Faure[1], L. S. I. Veiga[1], D. Meyers[4,5], S. G. Chiuzbaian[6,7], A. Nicolaou[6], N. Jaouen[6], R. B. Jackman[1], A. Nag[8], M. Garcia-Fernandez[8], Ke-Jin Zhou[8], A. C. Walters[8], K. Gilmore[4,9], D. F. McMorrow[1], M. P. M. Dean[4]

[1]University College London, London, United Kingdom. [2]Max Planck POSTECH/KOREA Research Initiative, Pohang, Korea, Republic of. [3]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany. [4]Brookhaven National Laboratory, Upton, USA. [5]Oklahoma State University, Stillwater, USA. [6]Synchrotron SOLEIL, Gif-sur-Yvette, France. [7]Sorbonne Universite, Paris, France. [8]Diamond Light Source, Didcot, United Kingdom. [9]Humboldt-Universitat zu Berlin, Berlin, Germany

Abstract

Interactions between electrons and phonons are important for a wide range of material properties, and techniques which can measure the strength of these interactions are highly sought-after. Recently, there has been considerable interest in the development of resonant inelastic x-ray scattering (RIXS) as a direct, momentum-resolved probe of electron-phonon (*e*-ph) interactions. Following seminal theoretical work by Ament *et al.*[1], RIXS studies of *e*-ph interactions have been conducted on a number of topical titanate[2], cuprate[3] and iridate[4] materials, among others.

In this talk, I will present carbon *K*-edge RIXS measurements of phonon excitations in graphite[5], chosen as a simple test material. By

tuning the incident x-ray energy to the π* and σ* resonances, I will demonstrate the ability of RIXS to probe the interaction between phonons and specific electronic states near to, and away from, the Fermi level respectively. Our high-resolution measurements reveal detailed structure in the multi-phonon features that directly encode the dispersion of the *e*-ph interaction strength. We extend a Green's-function method[6] to model this structure, which naturally accounts for the phonon and interaction-strength dispersions, as well as the mixing of phonon momenta in the intermediate RIXS state. This model reveals that the differences between the spectra at the two resonances can be fully explained by contrasting trends of the *e*-ph interaction through the Brillouin zone. Our results advance the interpretation of phonon excitations in RIXS and extend its applicability as a probe of *e*-ph interactions to a new range of out-of-equilibrium situations.

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Resonant x-ray studies in new generation Kitaev honeycomb iridates

Alberto De la Torre[1], Faranak Bahrami[2], Fazel Tafti[2], Kemp Plumb[1]
[1]Brown University, Providence, USA. [2]Boston College, Boston, USA

Abstract

The Kitaev Quantum spin liquid model can be realized in materials composed of d5 magnetic atoms in a strong octahedral crystal field and large spin-orbit coupling [1]. Of particular interest are Ir^{4+} with a network of edge-sharing IrO6 octahedra arranged in a honeycomb lattice, for example, α-Li_2IrO_3 and $NaIrO_3$, in which strong Kitaev interactions are present. However, the existence of finite Heisenberg and pseudo-dipolar interactions in these compounds leads to long-range magnetic order at low temperatures [2,3]. Aiming to modify the magnetic interactions in these compounds, new honeycomb iridates, including $Ag_3LiIr_2O_6$ [4] and $H_3LiIr_2O_6$ [5], have been synthesized by introducing inter-honeycomb-layers Ag and H. However, to date, there is little empirical understanding of the influence of the intercalated atoms on the electronic structure, local Ir environment, and magnetism in this new generation of Kitaev honeycomb iridates. In this talk, I will present a series of resonant x-ray spectroscopy measurements on a set of new-generation honeycomb iridates, including $Ag_3LiIr_2O_6$ [6] and $H_3LiIr_2O_6$ [7]. In the former, I will discuss the validity of the local jeff = ½ picture. In the latter, I will show evidence for dominant bond-direction interactions and a lack of short-range order in a Kitaev quantum spin liquid candidate.

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Electronic structure, magnetic interactions, and charge order in low valence nickelates probed by RIXS

Mark Dean, Yao Shen
Brookhaven National Laboratory, Upton, USA

Abstract

After a 30-year quest, researchers recently succeeded in realizing superconductivity in low valence nickelates [1]. This ignited a vigorous debate regarding the essential electronic properties of these materials and their similarity to cuprates. Some important questions include: Do these materials have appreciable oxygen charge-transfer character and superexchange akin to the cuprates or are they in a distinct Mott-Hubbard regime where oxygen plays a minimal role and superexchange is negligible? Given that cuprates have a propensity to host proximate competing phases such as charge and spin order, one might ask whether the nickelate's phase diagrams also host competing orders and to what extent they are similar to those in cuprates? In this talk, I will give a perspective of where we are with these questions, including our studies of the trilayer low valance nickelate $La_4Ni_3O_8$ [2,3].

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Resonant Inelastic X-ray Scattering as a Probe of Two-State Reactivity in Molecular Catalysis

Serena DeBeer
Max Planck Institute for Chemical Energy Conversion, Mülheim an der Ruhr, Germany

Abstract

Oxoiron(IV) intermediates are invoked as reactive intermediates in a wide range of heme and non-heme enzymes, which are capable of effecting selective substrate oxidation. In the case of non-heme enzymes, the characterized high-valent Fe(IV)-oxo complexes all have been shown to have quintet ground states. In contrast, the majority of the synthetic Fe(IV)-oxo complexes have triplet ground states. These molecular models are nevertheless still able to activate strong C-H bonds and computational studies suggest that this reactivity is enabled through so called “two-state reactivity”, where the complex crosses from a triplet ground state to a low-lying quintet in order to lower the energetic barrier for formation of high-spin products. Along the triplet surface, there are two potential pathways for hydrogen atom abstraction from substrate, the π-pathway where a β-electron is transferred into a singly occupied π* orbital, or the σ-pathway where the β-electron is transferred into the unoccupied σ*z orbital and relaxes down to a π* orbital. Along the quintet surface, there is only the σ-pathway available, where an α-electron is transferred to the σ*z orbital. Understanding the reactivity of these molecular complexes and of the enzymes requires a detailed understanding of the orbital energetics and more specifically the energy of low-lying excited states. Herein, recent 1s3p and 2p3d RIXS studies to assess to electronic structure of Fe(IV)-oxo catalysts will be presented. The relation of these results to the reactivity will be highlighted.

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Phonon quasiparticle breakdown and strongly anharmonic dynamics in energy materials: studies with inelastic scattering, diffuse scattering, and simulations

Olivier Delaire
Duke University, Durham, USA

Abstract

A deeper understanding atomic motions in solids is needed to refine microscopic theories of transport and thermodynamics, enabling the design of improved materials. In particular, the behavior of atomic vibrations (phonons) is key to rationalize numerous functional properties, ranging from fast ionic diffusion in solid-state electrolytes, to thermoelectrics for cooling or waste-heat harvesting, to photocarrier relaxation in photovoltaics. Yet, much remains to be understood about strong anharmonic effects in real materials. Strong anharmonicity, for instance near lattice instabilities, disrupts the conventional quasiharmonic phonon gas model and large vibrational amplitudes renormalize the electronic structure and electron-phonon interactions. This presentation will highlight our studies of strongly anharmonic atomic dynamics in several energy materials, including thermoelectrics [1,2], superionic conductors [3-6], and halide perovskite photovoltaics [7,8]. Our approach combines inelastic x-ray/neutron scattering with diffuse x-ray/neutron scattering to probe spatio-temporal correlations relevant to phonon propagation or solid-state diffusion. We integrate complementary first-principles and machine-learning accelerated simulations to enable the quantitative rationalization of our experiments, and identify underlying atomistic mechanisms.

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Inelastic X-ray Study of Elemental Metals Cerium (Ce) and Europium (Eu) at High Pressure

Yang Ding
Center for High-Pressure Science and Technology Advanced Research, Beijing, China

Abstract

Understanding the behaviors of 4f electrons is key to elucidating the paradigmatic physical phenomena such as valence fluctuation, insulator-metal, nonmagnetic-magnetic, and superconducting transitions in lanthanide metals and compounds, but it remains a long-standing challenge in many-body quantum physics. The use of external pressure to tune localized 4f states to delocalized ones offers a unique opportunity to explore the electrodynamics of 4f electrons. In a recent work, we applied inelastic x-ray scattering and resonant x-ray emission spectroscopy to investigate the changes of 4f states in the volume collapse transition of Ce [1] and valence transition of Eu [2] at high pressure, and our discoveries shed light on the underlying mechanism of the transitions.

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Low-energy spin dynamics in the diluted pyrochlore iridate $Tb_{2+x}Ir_{2-x}O_{7-y}$ (x = 0.3) probed by Resonant Inelastic X-rays Scattering

Quentin Faure[1], Anna Toschi[1], Elsa Lhotel[2], Dharmalingam Prabhakaran[3], Andrew Boothroyd[3], Desmond McMorrow[4], Christoph Sahle[1]
[1]European Synchrotron Radiation Facility, Grenoble, France. [2]Institut Neel, Grenoble, France. [3]Oxford University, Oxford, United Kingdom. [4]University College London, London, United Kingdom

Abstract

Low-energy spin dynamics in the diluted pyrochlore iridate $Tb_{2+x}Ir_{2-x}O_{7-y}$ (x = 0.3) probed by Resonant Inelastic X-rays Scattering

Quentin Faure[1], Anna Toschi[1], Elsa Lhotel[2], Dharmalingam Prabhakaran[3], Andrew Boothroyd[3], Desmond McMorrow[4] , Christoph Sahle[1]

1 European Synchrotron Radiation Facility, Grenoble, France

2 Institut Neel , Grenoble, France

3 University of Oxford, Clarendon Laboratory, United Kingdom

4University College London, London, United Kingdom

Iridate compounds have attracted a lot of attention in the past few years due to their strong spin-orbit coupling leading to a Jeff = 1/2 ground state that mixes both orbital and spin degrees of freedom [1,2,3].

Among those, the family of pyrochlore iridates RE2Ir2O7 (RE = Gd, Tb, Dy, Nd…) offers an interesting playground to explore this strong spin-orbit physics [1, 2] in addition to a weak-intermediate regime of Coulomb repulsion where novel topological phases have been theoretically predicted such as magnetic Weyl semi-metal states. Two conditions are necessary for realizing such states [1, 2]:

- The magnetic Ir sublattice must order in a “All In All Out” (AIAO) phase
- The electronic correlations must be in the “intermediate” regime

While the first condition is realized in most of the pyrochlore iridate family, the second is still subject to debate and recent studies now affirm that pyrochlore iridates might be in a strong correlations’ regime rather than a weak-intermediate regime necessary to realize semi-metallic states.

This work is devoted to the study of the low-energy spin-dynamics of the diluted (non-stoichiometric) compound Tb(2+x)Ir(2-x)O7-y (x = 0.3). Besides the fact that the spin-dynamics of only few members of the pyrochlore iridates’ family have been investigated [4,5], the motivation of this study lies into two points. First, recent resistivity measurements showed that magnetic dilution lowers the electronic correlations. Second, the spin-dynamics response from a magnetic system is closely linked to either its itinerant or localized character. Hence the study of the spin dynamics could be considered as an indirect way to elucidate the strength of the electronic correlations in pyrochlore iridates [6].
We hereby expose recent Resonant Inelastic X-rays Scattering (RIXS) measurements that allowed us to probe the spin-dynamics of this compound along high-symmetry directions of the Brillouin zone. In

addition, we performed spin-waves calculations using spinW software that allowed us to extract the microscopic parameters of the low-energy Hamiltonian. Finally, some discrepancies between experimental results and a strong correlations approach assumed in spin-waves calculations are observed that could be indicative of weaker correlations in this compound.

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Email corresponding author: quentin.faure@esrf.fr

Prefered presentation – Poster

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How cuts through the manifold of molecular H_2O potential energy surfaces from RIXS back a continuous distribution models of liquid water at ambient conditions

Alexander Föhlisch
Potsdam University, Potsdam, Germany. Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany

Abstract

The phase diagram of water harbors controversial views on underlying structural properties of its constituting molecular moieties, its fluctuating hydrogen-bonding network, as well as pair-correlation functions. We establishe with vibrationally resolved RIXS the manifold of molecular H_2O potential energy surfaces in liquid water at ambient conditions in direct comparison to the gas phase molecule [1,2,3,4,5,6]. A broad approach of quantitative and high-resolution multimethod X-ray spectroscopic investigation [7] uses long energy-range X-ray absorption to calibrate data for water gas, liquid, and ice to the experimental atomic ionization cross-section. For liquid water, the mean value of 1.74 ± 2.1% donated and accepted hydrogen bonds per molecule is extracted, pointing towards a continuous-distribution model for liquid water at ambient conditions as do X-ray emission spectra once femtosecond scattering duration and proton dynamics are taken into account [7].

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36

Single crystal elasticity at extreme conditions

Hiroshi Fukui
JASRI, Sayo, Japan

Abstract

We have an extensive program measuring the elastic properties of single crystal in extreme, e.g. high pressure, conditions using x-ray scattering because of the high penetrability of hard x-ray. This is based on inelastic x-ray scattering and an analysis with the Christoffel equation. It has the advantage of using both symmetry and off-symmetry directions and a 2D analyzer array making measurements both precise and efficient [1]. Simultaneous measurement of a molar volume and an elasticity of a material yields an equation of states (EoS) of the material independently to other pressure standards by numerically integration of a bulk modulus. We have measured single crystal elasticity of pressure-standard materials and demonstrated to establish absolute pressure scales [2-5]. We review how to establish EoSs based on single crystal elasticity.

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21

The manyfold excitations of cuprates and nickelates observed via high resolution RIXS

Giacomo Ghiringhelli
Politecnico di Milano, Milano, Italy

Abstract

The rich physics of cuprates is very effectively captured by high resolution RIXS experiments made at Cu L_3 and O K edges. This fortunate conjuncture has boosted the development of better and better instrumentation at synchrotrons and has served as one of the scientific cases for RIXS at XFELs. The field is expanding and experiments are leading to more insightful results, where the different degrees of freedom are organically studied.

I will provide a survey of our recent work on cuprate parent compounds and superconductors and on infinite layer nickelates, which share several properties with high Tc superconductors. I will present mostly unpublished results on fractionalized spin excitations [1], dispersing orbital excitations, charge order [2] and charge density fluctuations, doping dependent electron phonon coupling [3] and T-dependent low energy charge excitations, in various compounds including YBCO, $CaCuO_2$ and $Nd_{1-x}Sr_xNiO_2$. I will also mention the very first pump probe RIXS experiments on cuprates and other quantum materials at the European XFEL.

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electron-phonon coupling in two families of bilayer superconducting cuprates. Phys. Rev. B 105, 115105 (2022).

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Characterization of Metal-Centered Excited States of Cobalamin Using 2p3d RIXS Spectroscopy

Nahid Ghodrati[1], Rebeca G. Castillo[2], Benjamin E. Van Kuiken[1]

[1]European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany.

[2]Laboratory for Ultrafast X-ray Sciences, École polytechnique fédérale de Lausanne, Lausanne CH-1015, Swaziland

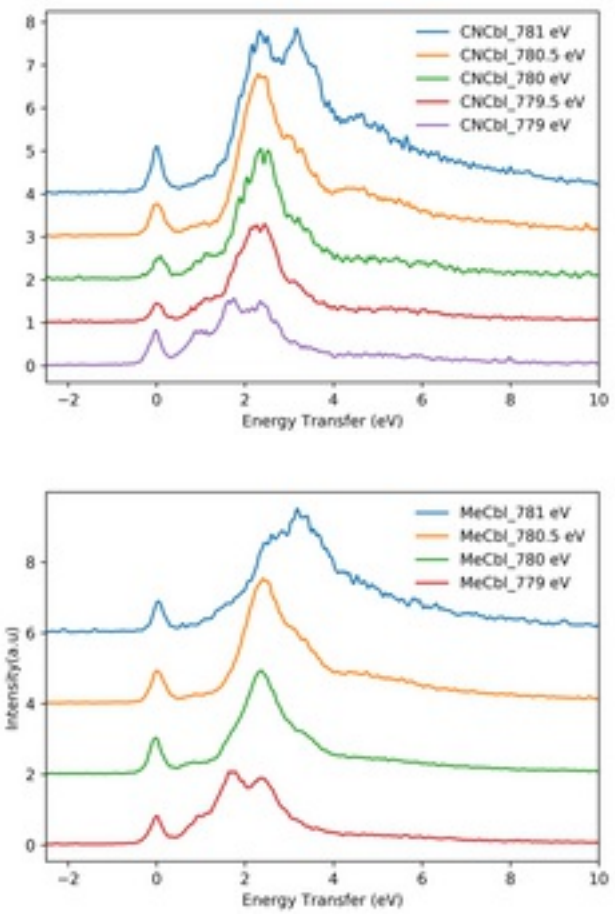
Abstract

Cobalamins are complex organometallic and biological cofactors that play a crucial role in human metabolism.[1] Although many years have passed since the initial isolation of crystalline vitamin B_{12}[2], recent advances in the field of X-ray spectroscopy have prompted further detailed studies on their chemical dynamics.

Photoexcitation with green light in ultrafast dynamics is characterized by a cascade through a series of electron excited states.[3] It has been proposed that the initial ππ* excited state transitions into a ligand to metal charge transfer (LMCT) state or metal-centered excited state. However, these transitions have not been spectroscopically probed yet. The investigation of these transitions requires the use of spectroscopic techniques that are element specific and sensitive to changes in the local electronic structure. In this work 2p3d RIXS measurements at the Co L-edge are used to valence electronic structure in cobalamins.

The RIXS spectra for cyanocobalamin (CNCbl) and methylcobalamin (MeCbl) were collected across the L_3-edge. These samples differ in the identity of the axial ligand in their structures. The information acquired from 2p3d RIXS experiments reveal that d-d spectra are different due to changes in axial ligands (Fig.1a). This study focuses on the experimental methods used to acquire the data and computational methods used for analysis.

a


CNCbl_781 eV
CNCbl_780.5 eV
CNCbl_780 eV
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CNCbl_779 eV
Energy Transfer (eV)
MeCbl_781 eV
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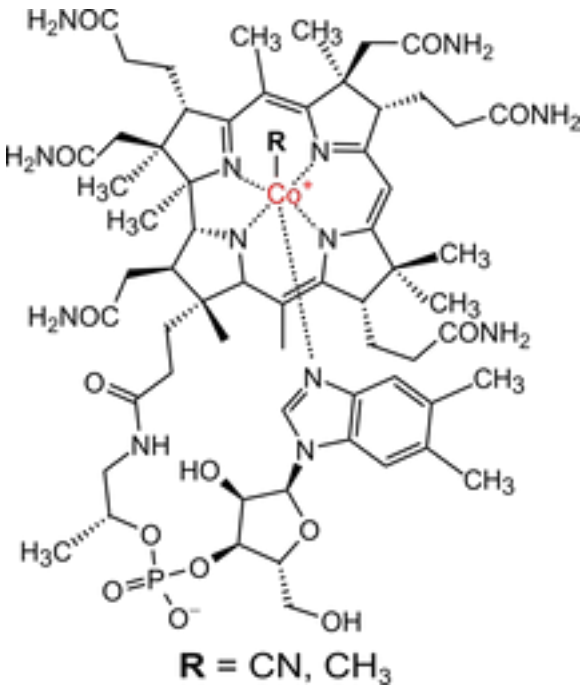
MeCbl_779 eV
Intensity(a.u)
Energy Transfer (eV)

b


H2NOC
CH3 CH3
CONH2
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R
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H3C
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N
N
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CH3
H2NOC
CONH2
O
N
CH3
NH
N
CH3
HO
H3C
O
O
O=P-O
O⁻
OH
R = CN, CH3

Figure 1. ***(a)*** The 2p3d RIXS spectra cascade of the CNCbl, MeCbl, and ***(b)*** their structures.

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Email corresponding author: Nahid.ghodrati@xfel.eu

Prefered presentation – Poster

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Description of resonant inelastic x-ray scattering in correlated metals

Keith Gilmore
Humboldt University, Berlin, Germany

Abstract

Resonant inelastic x-ray scattering (RIXS) provides unprecedented experimental access to the fundamental excitations and dynamical responses of correlated materials. When RIXS is applied to insulating or weakly metallic correlated materials the resulting complex spectral signatures are amenable to interpretation by numerical techniques that restrict the active subspace. Correlated metals, with their increased bandwidth and hybridization, remain beyond the feasibility limits of such computational methods. We present new, first-principles methodology for calculating the RIXS response of correlated metals in an unbiased fashion and without restriction to particular orbital subsets [1]. This begins with a Bethe-Salpeter equation (BSE) calculation that gives the quasiparticle contribution to the RIXS spectrum. We augment this by generating secondary bosonic excitations through a real-time time-dependent density functional theory calculation. By combining these two contributions we are able to accurately account for both direct and indirect RIXS features using parameter-free first-principles calculations. The approach is amenable to both transition metal L-edges and ligand K-edges. We validate our new methodology by accurately reproducing the experimental RIXS response at the Fe L_3 edge of $BaFe_2As_2$ and the V L_3 edge of $SrVO_3$.

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Photon-in/photon-out spectroscopy in materials science

Pieter Glatzel
ESRF, Grenoble, France

Abstract

Instruments for photon-in/photon-out spectroscopy have become available on many X-ray absorption spectroscopy beamlines at synchrotron radiation sources. This makes the technique visible and accessible to many researchers in the materials science community. In particular, in situ and operando studies in catalysis and research related to energy storage greatly benefit from the larger information content when X-ray emission spectroscopy (XES) is added to X-ray absorption spectroscopy. Beamline ID26 at the ESRF features XES instruments for the hard and tender X-ray range. Their design maximizes detection efficiency for low analyte concentrations and time-resolved studies. In situ cells for tender X-rays must be inserted into the vacuum of the spectrometer chamber and windows have to be adapted to the short penetration depth of the tender X-rays. I will present the tender X-ray spectrometer and some examples for the application of photon-in/photon-out spectroscopy in materials science. I will furthermore discuss a new protocol for time-resolved and imaging studies using X-ray emission spectroscopy.

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Bond-directional magnetic excitations in Kitaev materials studied by RIXS

Markus Grüninger
University of Cologne, Cologne, Germany

Abstract

The concept of a 'proximate spin liquid' was coined in the context of the Kitaev materials Na_2IrO_3 and α-$RuCl_3$. These materials exhibit magnetic order at low temperatures - though of spin-orbit entangled j=1/2 moments that are subject to dominant bond-directional Kitaev exchange, which has made them candidates for the sought-after Kitaev quantum spin liquid with fractional Majorana excitations. The idea is that above the magnetic ordering temperature there exists an extended regime in which aspects of the Kitaev spin liquid such as the fractional nature of excitations may still be realized – the proximate spin liquid.

As an experimental probe of this scenario, we study RIXS at the Ir *L* edge in the honeycomb iridates. We observe distinctive fingerprints of Kitaev physics [1], in particular an excitation continuum that persists up to at least 300 K. The corresponding dynamical spin-spin correlations are restricted to nearest neighbors, a key property of the Kitaev model. To explicitly probe the bond-directional character of excitations is an experimental challenge since it requires to *simultaneously* determine the spin operator that creates an excitation as well as the direction of the bond involved. We show that RIXS is ideally suited to address this demanding task, allowing us to derive a comprehensive picture of the unusual magnetic excitations.

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Unveiling the nature of oxidised oxygen in battery cathode materials with RIXS

Robert House
University of Oxford, Oxford, United Kingdom

Abstract

The energy density of Li-ion batteries can be improved by storing charge at high voltages through the oxidation of oxide ions in the cathode material. However, oxidation of O^{2-} triggers irreversible structural rearrangements in the bulk and loss of the high voltage plateau on charge, which is replaced by a lower discharge voltage. Understanding the nature of oxidized oxygen has proved a significant experimental challenge.

We used high resolution RIXS at the O K-edge to reveal the electronic changes that occur on oxygen during the oxidation and reduction reactions. At an excitation energy of 531 eV, a series of sharp energy loss peaks can be resolved in the charged materials which arise from transitions to vibrationally excited states of O_2 molecules. The O_2 is trapped within the structure and can be reduced on discharge back to O^{2-}, but this process occurs at a lower voltage compared to the first charge, giving rise to voltage hysteresis. We show that O_2 formation is observed across a range of different structures and compositions and demonstrate the power of RIXS to distinguish between O_2, O_2^{-} and O_2^{2-} species in the bulk of solids.

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Exciton excitation of superconducting cuprates studied by soft X-ray RIXS

A. Singh[1], H. Y. Huang[1], J. D. Xie[2], J. Okamoto[1], C. T. Chen[1], T. Watanabe[3], A. Fujimori[4], M. Imada[5], Di-Jing Huang[1]
[1]NSRRC, Hsinchu, Taiwan. [2]National Yang Ming Chiao Tung University, Hsinchu, Taiwan. [3]Hirosaki University, Hirosaki, Japan. [4]University of Tokyo, Tokyo, Japan. [5]Waseda University, Tokyo, Japan

Abstract

Electron quasiparticles play a crucial role in simplifying the description of many-body physics in solids with surprising success. Conventional Landau's Fermi-liquid and quasiparticle theories for high-temperature superconducting cuprates have, however, received skepticism from various angles. A path-breaking framework of electron fractionalization has been established to replace the Fermi-liquid theory for systems that show the fractional quantum Hall effect and the Mott insulating phenomena; whether it captures the essential physics of the pseudogap and superconducting phases of cuprates is still an open issue. Here, we show that excitonic excitation of optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ with energy far above the superconducting-gap energy scale, about 1 eV or even higher, is unusually enhanced by the onset of superconductivity. Our finding proves the involvement of such high-energy excitons in superconductivity. Therefore, the observed enhancement in the spectral weight of excitons imposes a crucial constraint on theories for the pseudogap and superconducting mechanisms. A simple two-component fermion model which embodies electron fractionalization in the pseudogap state well explains the change, pointing toward a novel route for understanding the electronic structure of superconducting cuprates.

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Quantum Fluctuations of Charge Order Induce Phonon Softening in $La_{0.5}Sr_{1.5}CuO_4$

Hsiao-Yu Huang[1], Amol Singh[1], Chung-Yu Mou[2], Steve Johnston[3], Alexander F. Kemper[4], Jeroen van den Brink[5], Peng-Jen Chen[6,7], Ting-Kuo Lee[8,9], Jun Okamoto[1], Yen-Yi Chu[1], Jung-Han Li[10], Seiki Komiya[11], Alexander Komarek[12], Atsushi Fujimori[1,10,13], Chien-Te Chen[1], Di-Jing Huang[1,10]

[1]National Synchrotron Radiation Research Center, Hsinchu, Taiwan. [2]Center for Quantum Technology and Department of Physics, National Tsing Hua University, Hsinchu, Taiwan. [3]Department of Physics and Astronomy, The University of Tennessee, Knoxville, Tennessee, USA. [4]Department of Physics, North Carolina State University, Raleigh, North Carolina, USA. [5]nstitute for Theoretical Solid State Physics, IFW Dresden, Dresden, Germany. [6]Department of Mechanical Engineering, City University of Hong Kong, Hong Kong, China. [7]Hong Kong Institute for Advanced Study, City University of Hong Kong, Hong Kong, China. [8]Institute of Physics, Academia Sinica, Taipei, Taiwan. [9]Department of Physics, National Sun Yat-sen University, Taipei, Taiwan. [10]Department of Physics, National Tsing Hua University, Hsinchu, Taiwan. [11]Central Research Institute of Electric Power Industry, Yokosuka, Kanagawa, Japan. [12]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany. [13]Department of Physics, University of Tokyo, Tokyo, Japan

Abstract

Quantum phase transitions play an important role in shaping the phase diagram of high-temperature cuprate superconductors. These cuprates possess intertwined orders which interact strongly with superconductivity. Recently, charge charge-density waves (CDWs) in

cuprate superconductors have attracted renewed interest. However, the evidence for the quantum critical point associated with the charge order in the superconducting phase remains elusive. Here, we reveal the short-range charge orders and the spectral signature of the quantum fluctuations in $La_{2-x}Sr_xCuO_4$ (LSCO) near the optimal doping using high-resolution resonant inelastic x-ray scattering. On performing calculations through a diagrammatic framework, we discover that the charge correlations significantly soften several branches of phonons. The quantum critical scalings of charge fluctuations in the temperature domain are clearly demonstrated, consistent with the speculation on the existence of a QCP. These results elucidate the role of charge order in the LSCO compound, providing evidence for quantum critical scaling and discommensurations associated with charge order.

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Probing 3d-4s orbital hybridisation of linear transition metal complexes through Resonant Inelastic X-ray Scattering

Myron Huzan[1,2], Manuel Fix[3], Anton Jesche[3], Michael Baker[1,2]
[1]University of Manchester, Manchester, United Kingdom. [2]University of Manchester at Harwell, Oxford, United Kingdom. [3]University of Augsburg, Augsburg, Germany

Abstract

Low coordinate single-ion magnets (SIMs) have attracted much attention due to exhibiting exceptional energy barriers for reversal of magnetism which would be of benefit to information storage and quantum information processing applications [1]. Linearly coordinated transition metal complexes facilitate the first-order spin-orbit coupled ground states, that are otherwise typically quenched in transition metal sites through a Jahn-Teller distortion. This enables a large easy-axis magnetic anisotropy due to the presence of a bistable ground state separated by an energy barrier to magnetisation reversal. The synthesis of linear transition metal SIMs has advanced with recent developments of novel complexes possessing unique orbital ordering and valences in the cooperative pursuit of rare-earth alternative high-temperature single ion anisotropic magnets. However, the characterisation of these complexes has been limited to theoretical calculations.

The doping of metal ions within extended crystalline solids provides an alternative method to explore single-ion magnetism relative to coordination chemistry approaches. Lithium nitride is proven to be an excellent host for studying the single-ion magnetism of dopants [2,3]. Fe doped lithium nitride has been demonstrated to be a remarkable single-ion-magnet, exhibiting a magnetic coercivity field exceeding many rare-earth-based permanent magnets with a

hysteresis that persists up to 16 K and relaxation rates on the order of ~104 s [4]. In this study, single-crystal angular dependent L_3-edge RIXS maps are semi-empirically characterised through atomic multiplet calculations within Quanty [5] with insights and comparison to ab initio CASSCF calculations. Quantification of the ligand-field and spin-orbit coupling of this complex is discussed, and the significance of 3d-4s hybridisation and metal-to-ligand charge transfer.

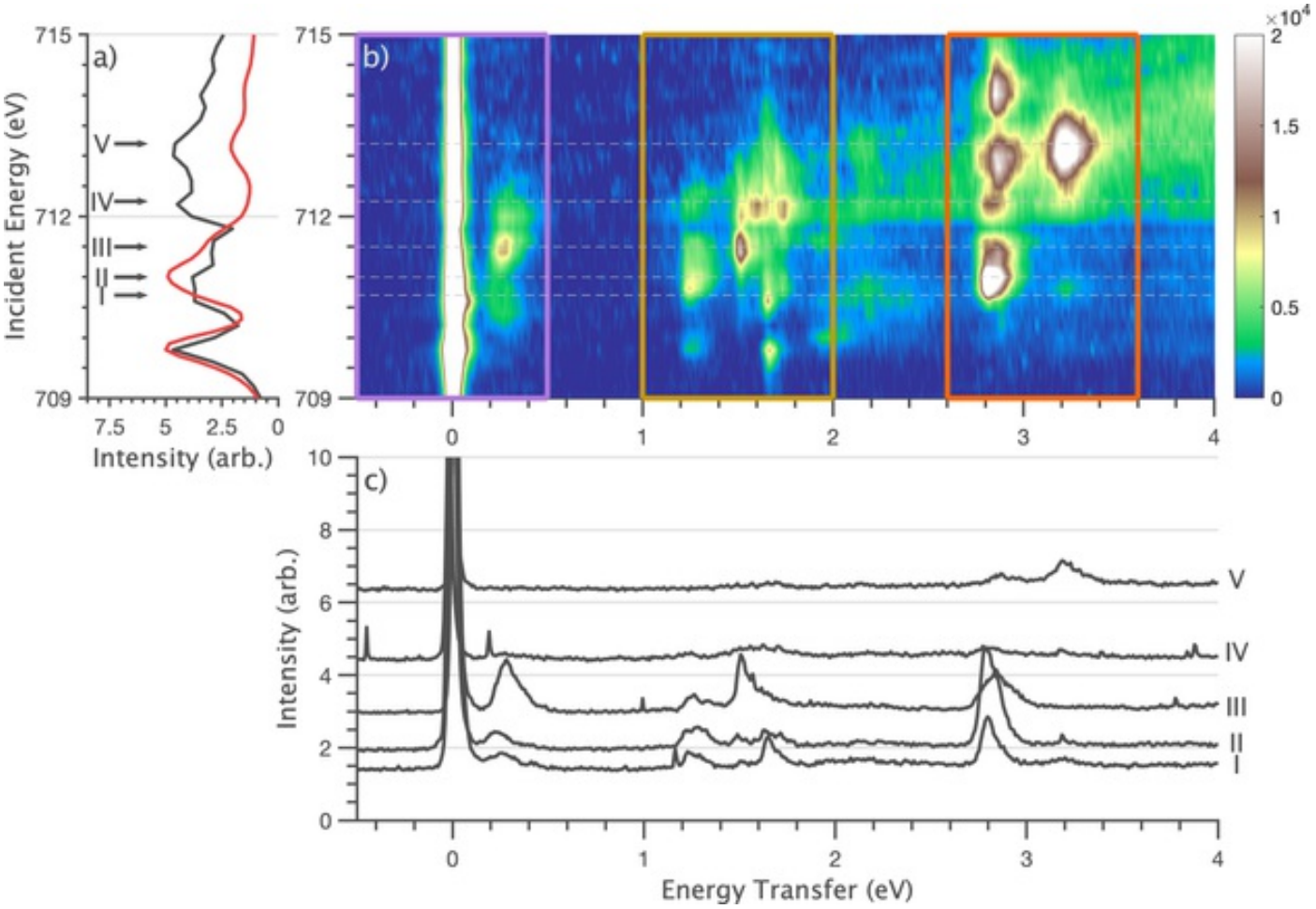

Figure 1: a) Comparison of L_3-edge $Li_2(Li_{1-x}Fe_x)N$ XAS spectra of integrated energy transfer RIXS (black) and FY-XAS (red). b) Fe L_3-edge RIXS map with incident X-ray vector, k_i // c performed at 11K with nominal concentration of x = 0.01. c) Several constant incident energy RIXS cuts highlighting discrete spectral features and relative intensities corresponding to various orbital excitations.

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Title: Resonant inelastic x-ray scattering as a probe of multipolar and nonlocal magnetic excitations.

Speaker: Steven Johnston, University of Tennessee, Knoxville

Abstract: Resonant inelastic x-ray scattering is a powerful probe of strongly correlated quantum materials, with combined access to collective spin, charge, orbital, and lattice excitations. Notably, we have recently demonstrated that RIXS can access novel multipolar magnetic excitations due to the presence of the core hole and its corresponding dynamics in the intermediate state of the scattering process. Examples include higher-order four-spinon and quadrupolar magnetic excitations in one-dimensional antiferromagnetic spin-1/2 and spin-1 chains. In this talk, I will discuss the theoretical perspective of these studies, including how these excitations can provide information on nonlocal spin-spin correlations. I will also discuss the potential applications of these measurements for determining core-hole lifetimes in the context of probing electron-lattice couplings with RIXS.

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X-ray Raman Spectroscopy instrumentation at the EMA beamline

Ulisses Kaneko[1], José Corsaletti Filho[1], Marcos Eleotério[1], Joel Mendonça[1], Guilherme Calligaris[1], Audrey Grockowiak[1], Ricardo Reis[1], Diego Casa[2], Narcizo Souza-Neto[1]

[1]Brazilian Synchrotron Light Laboratory, Campinas, Brazil.

[2]Advanced Photon Source, Lemont, USA

Abstract

The Extreme Method of Analysis (EMA) is a beamline of the Brazilian synchrotron, Sirius, designed to apply synchrotron light in materials subjected to extreme conditions of pressure, temperature, and magnetic fields [1]. At the EMA beamline, the users can run X-ray diffraction (XRD) to X-ray absorption (XAS and XMCD) experiments to study a broad range of compounds with submicrometric beamsize and energy between 3 – 30 keV. At the end of the hutch, single crystals can be studied through high-resolution XRD in a 6+2 circle diffractometer and at the same instrument, the inelastic X-ray spectrum of light elements can be measured with hard X-rays through X-ray Raman spectroscopy (XRS). In this talk, I will show details and main parameters of the XRS instrument in commissioning at the EMA beamline that is installed at the 2θ arm of the diffractometer. This configuration allows for forward, backscattering and momentum dependent XRS spectroscopy. It will be discussed the manufacturing process of the Si analyzers, the control of their positioning modules in the alignment procedure using picomotors, and how we process images to produce an XRS spectrum. Finally, the energy resolution obtained with different diced Si analyzers will be compared and the first results at the K-edge of C of graphite and diamond will be shown.

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RIXS from 4d-electron compounds

Bernhard Keimer
Max Planck Institute for Solid State Research, Stuttgart, Germany

Abstract

Intermediate-energy resonant inelastic x-ray scattering (IRIXS) with photon energies in the “tender” x-ray regime (2.5-5 keV) has recently been established at beamline P01 at the PETRA-III synchrotron [1,2]. The energy resolution of the IRIXS instrument is currently ~30 meV at the Ru L-edge, comparable to the most advanced soft x-ray RIXS instruments. We will highlight recent experiments on collective spin excitations as well as spin-state transitions and spin-orbit excitons in a variety of ruthenium compounds [3-8], and outline perspectives for future IRIXS studies of 4d-electron compounds and heterostrucures. We will also compare and contrast these results with neutron scattering data on related compounds [9,10].

The work discussed in this talk is based on collaboration with many scientists, who are listed in the publications below. Financial support from the European Research Council Advanced Grant 669550 (Com4Com) is gratefully acknowledged.

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RIXS investigation of Kitaev materials

Blair Lebert[1], Subin Kim[1], Felix Frontini[1], SaeHwan Chun[2], Kejin Zhou[3], Mirian Garcia-Fernandez[3], Stefano Agrestini[3], Jaewon Choi[3], Diego Casa[4], Young-June Kim[1]

[1]University of Toronto, Toronto, Canada. [2]Pohang Light Source, Pohang, Korea, Republic of. [3]Diamond Light Source, Didcot, United Kingdom. [4]Advanced Photon Source, Argonne, USA

Abstract

Magnetic materials with 4d or 5d transition metals have drawn much attention for their unique magnetic properties arising from J_eff=1/2 magnetic states. Among them, a honeycomb lattice material with unusual bond-dependent interactions called Kitaev interactions is of particular interest due to the potential for realizing the Kitaev quantum spin liquid state. Although much progress has been made in understanding magnetic and spin-orbit excitations in Kitaev materials, such as Na2IrO3 and alpha-RuCl3, using resonant inelastic X-ray scattering (RIXS), there are still many unanswered questions regarding the nature of electronic excitations in these materials. Of particular interest is the sharp peak observed around 0.4 eV in the RIXS spectrum of Na2IrO3 [1], the exact nature of which remains controversial. In this context, it is interesting to note that a similar lower energy “excitonic” peak was observed in alpha-RuCl3, in the Ru L3 edge RIXS investigation of Suzuki et al. [2]. Given that the electronic hopping parameters in alpha-RuCl3 are probably very different from those in Na2IrO3 (alpha-RuCl3 has a large bandgap of ~1eV, well above any SO excitation energy scale), the observed similarity is somewhat surprising. To understand the origin of this exciton peak, we have investigated the momentum and temperature dependence of the 0.4 eV peak in another Kitaev material, alpha-Li2IrO3. In addition, we have carried out a high-resolution M3-edge RIXS investigation of alpha-RuCl3 to study the exciton peak in detail. The RIXS spectra from these three compounds will be compared and

the origin of the exciton peak will be discussed in the context of recent theoretical studies.

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2p3d and 3p3d RIXS of NiO: A Multiplet Calculation Study

Masoud Lazemi[1], Uwe Bergmann[2], Frank de Groot[1]
[1]Utrecht University, Utrecht, Netherlands. [2]University of Wisconsin-Madison, Madison, USA

Abstract

X-ray absorption spectroscopy (XAS) and resonant inelastic X-ray scattering (RIXS) have garnered significant attention due to their ascendancy in revealing the geometric structure, chemical bonding, charge, and spin states in addition to chemical sensitivity. Hitherto, K-edge XAS (1s→3d/4p) and RIXS (1s2p), and L-edge XAS (2p→3d) and RIXS (2p3d) of 3d transition-metal complexes have been employed extensively. However, despite the seminal work by Kuiper et al.[1] in 1998, limited studies have used the M-edges corresponding to 3p→3d transitions below 100 eV, e.g., Fe at 52.7 eV and Ni at 66.2 eV.[2] Recently, with the advent of lab-based ultrafast extreme ultraviolet (XUV) sources, this spectral range is starting to gain some momentum.[3–5] On the excitation energy axis, the RIXS spectra give evidence of magnon and bi-magnon excitations,[6] in agreement with the theoretical spectra from 1998.[7]

Herein, we present L-edge (2p3d) and M-edge (3p3d) RIXS calculations of NiO based on multiplet theory[8] (Fig. 1a and 1b). The NiO has a Ni^{2+} ($3d^8$) configuration and cubic symmetry, which has the lowest number of multiplet states for any S>1/2 system.[6] Moreover, the associated energies of these states are already known. As a strongly correlated antiferromagnetic insulator, NiO has an effective interatomic superexchange interaction of 110 meV. This is sufficient to segregate the single (100 meV) and double spin-flip excitations (200 meV), shown in the bottom panels in Fig. 1 (a) and 1(b). The improved resolution will make the magnons visible also at

the Ni M-edge.

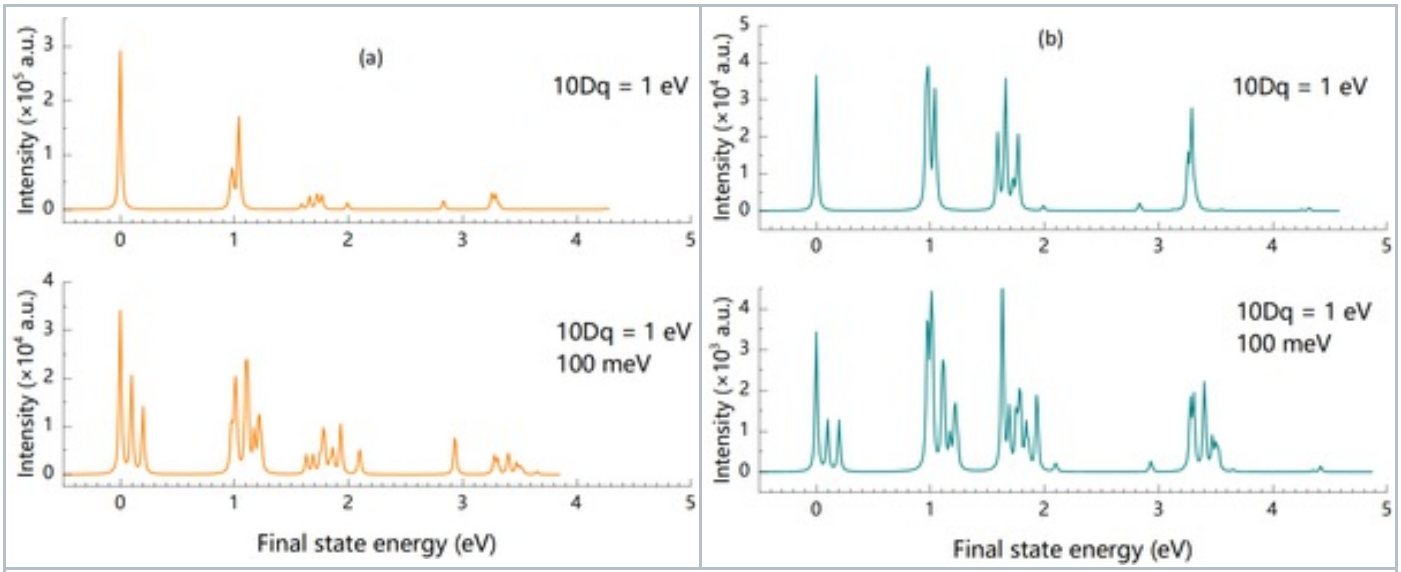

Fig. 1. (a) and (b) 2p3d and 3p3d RIXS of NiO, respectively. Upper panels: 10Dq = 1 eV, Bottom panels: 10Dq = 1 eV and 100 meV splitting.

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Soft-phonon and charge-density-wave formation in nematic BaNi2As2

Sofia-Michaela Souliou[1], Tom Lacmann[1], Rolf Heid[1], Christoph Meingast[1], Luigi Paolasini[2], Amir-Abbas Haghighirad[1], Michael Merz[1], Alexey Bosak[2], Matthieu Le Tacon[1]
[1]IQMT - KIT, Karlsruhe, Germany. [2]ESRF, Grenoble, France

Abstract

We use diffuse and inelastic x-ray scattering to study the formation of an incommensurate charge-density-wave order (I-CDW) in BaNi2As2, a candidate system for charge-driven electronic nematicity. At low temperatures, the I-CDW sets in before a structural transition to a triclinic phase, within which it is suppressed and replaced by a commensurate CDW order (C-CDW). Intense diffuse scattering signal is observed around the modulation vector of the I-CDW, QI−CDW already visible at room temperature and collapsing into superstructure reflections in the ordered state. A clear dip in the dispersion of a low-energy transverse optical phonon mode is observed around QI−CDW. The phonon continuously softens upon cooling, ultimately driving the transition to the I-CDW state. The transverse character of the soft-phonon branch elucidates the complex pattern of the I-CDW satellites and settles the debated unidirectional nature of the I-CDW. The phonon instability and its reciprocal space position is well captured by our ab initio calculations. These however indicate that neither Fermi surface nesting, nor enhanced momentum-dependent electron-phonon coupling can account for the I-CDW formation, demonstrating its unconventional nature.

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Signatures of Strong Electronic Correlations in Infinite-layer Nickelates

Wei-Sheng Lee
SLAC National Accelerator Lab., Palo Alto, USA

Abstract

The recent discovery of superconducting nickelates has drawn significant interest in the field. The nickelate superconductors are hole-doped infinite-layer nickelates $R\mathrm{NiO}_2$, which are isostructural to the infinite-layer cuprates and possesses the same nominal 3d electron count. Experiments and theories have revealed some notably differences to the cuprates, such as being a multi-orbital system instead of a single-band system; yet, whether strong electronic correlations are also at play is an important question to answer.

In this talk, I will address this issue by presenting our recent RIXS studies on infinite-layer nickelates. We have identified a branch of dispersive magnetic excitations reminiscent of the spin wave of a spin ½ AFM square lattice system in the undoped compounds. The high band width (200 meV) of the magnetic excitations reflects a relatively strong onsite Coulomb interaction. In addition, we have also revealed a charge order that exists in a notable portion of the phase diagram, indicative of a possible interplay between superconductivity, magnetism, and charge ordering. These observations are key signatures of strongly electronic correlations, which should play an important role in determining the electronic properties of the nickelate superconductors.

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Bond-directional nearest-neighbor excitations in the proximate Kitaev spin liquid Na_2IrO_3 probed by RIXS

Marco Magnaterra[1], Alessandro Revelli[1], Karolin Hopfer[1], Christoph Sahle[2], Marco Moretti Sala[3], Giulio Monaco[4], Jan Attig[5], Luca Peterlini[5], Ciarán Hickey[5], Petra Becker[6], Ladislav Bohatý[6], Anton Jesche[7], Philipp Gegenwart[7], Simon Trebst[5], Paul H. M. van Loosdrecht[1], Jeroen van den Brink[8], Markus Grüninger[1]
[1]Institute of Physics II, University of Cologne, Cologne, Germany. [2]European Synchrotron Radiation Facility, Grenoble, France. [3]Dip. di Fisica, Politecnico di Milano, Milano, Italy. [4]Dip. di Fisica, Università di Padova, Padova, Italy. [5]Institute of Theoretical Physics, University of Cologne, Cologne, Germany. [6]Institute of Geology and Mineralogy, Cologne, Germany. [7]Exp. Physics VI, University of Augsburg, Augsburg, Germany. [8]Inst. for Theo. Solid State Physics, IFW Dresden, Dresden, Germany

Abstract

The Kitaev model hosts a spin-liquid ground state with Majorana fermion excitations. It is based on bond-directional exchange, i.e. Ising-like interactions that couple different spin components on different bonds.

In Na_2IrO_3, resonant inelastic x-ray scattering (RIXS) revealed fingerprints of Kitaev physics in the magnetic excitations [1]. In fact, the RIXS intensity shows a sinusoidal **q** dependence that proves the nearest-neighbor or single-bond character of the excitations.

We report on refined RIXS measurements where we exploit the polarization dependence of the different spin channels and the **q** dependence of the different bonds to demonstrate the direct

connection between spin component and bond direction.

Our results establish the bond-directional nearest-neighbor character of magnetic excitations in Na_2IrO_3.

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Time-resolved *in situ* XAS study towards a Mechanistic Understanding of the Growth of Copper Nanoparticles

Sushila Marlow, Ryan Wang
University College London, London, United Kingdom

Abstract

Nanoparticles are widely used in catalysis where their surface properties, such as shape and size, have a strong effect on their behaviour. Their enhanced properties make them versatile, underlining the importance of work towards understanding and predicting the growth, rather than current trial-and-error methods. In this work, high-time resolution millisecond-scale XANES and EXAFS data is collected for the reduction of copper acetate under various reaction conditions. Valence-to-core XES and EPR are used supplement this study, to probe the chemistry of the reaction intermediates. By confirming intermediates, the coordination changes in EXAFS can be fitted for real-time growth data. The Cu reduction pathway in the presence of oxygen was successfully observed, which leads to a Cu^{1+} intermediate. Conversely, as expected, in an oxygen free environment, no Cu^{1+} was formed. EPR confirms the presence of a $Cu(N_2H_4)_2^{2+}$ coordinated intermediate. We are also able to monitor with high resolution the rates of reduction of Cu^{2+} and Cu^{1+} into Cu^0, which will later used to confirm a mechanistic model and predict nanoparticles formed under different temperature and concentration conditions.

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Spin and orbital excitations beyond the nearest neighbor interactions: spinon pairs and dispersing dds in infinite-layer $CaCuO_2$

Leonardo Martinelli[1], Davide Betto[2], Kurt Kummer[2], Riccardo Arpaia[3], Daniele Di Castro[4], Marco Moretti Sala[1], Nicholas B. Brookes[2], Thomas P. Devereaux[5], Lucio Braicovich[1], Giacomo Ghiringhelli[1]

[1]Dipartimento di Fisica, Politecnico di Milano, I-20133 Milano, Italy. [2]ESRF - The European Synchrotron, FR-38043 Grenoble, France. [3]Quantum Device Physics Laboratory, MC2 Department, Chalmers University, SE-41296 Goteborg, Sweden. [4]Dipartimento di Ingegneria Civile e Ingegneria Informatica, Università di Roma Tor Vergata, I-00133 Roma, Italy. [5]Stanford Institute for Materials and Energy Sciences, SLAC National Laboratory and Stanford University, 94025 - Menlo Park, USA

Abstract

The building block of cuprate high-T_c superconductors are CuO_2 planes where, in the absence of charge doping, the spin ½ sites interact via a huge antiferromagnetic exchange. The resulting ground state has (x^2-y^2) ferro-orbital symmetry and Néel antiferromagnetic order. The corresponding elementary excitations are commonly well described as local (non-collective) dd excitations, which show no dispersion with momentum, and ΔS=1 magnons (plus multi-magnons), respectively. However, longer range interactions cannot always be neglected. We have performed Resonant Inelastic X-ray Scattering (RIXS) measurements on the infinite-layer compound $CaCuO_2$, where the absence of apical oxygens leads to larger than usual hopping integrals t and t', superexchange J, and ring-exchange J_c [1]. In the orbital spectrum we find dispersive orbital excitations and the lifting of the *xz/yz* degeneracy, which

indicates a partially delocalized nature. These results are reminiscent of what happens in 1-dimensional spin chains [2] and in two-dimensional iridates [3]: interestingly, however, we observe a dispersion with a qualitative different shape. Even more interestingly, the magnetic spectral region shows clear anomalies close to (1/2,0), where the spectral weight of the magnon peak decreases and another higher-energy excitation takes the majority of the spectral weight. In order to unravel the nature of this anomaly, we employed an innovative combination of RIXS measurements at the Copper L3 edge. We have acquired polarization-resolved RIXS spectra with high resolution (40 meV) in different geometries, detuning spectra, and selected scans with ultra-high resolution (26 meV) to fully disentangle the different features of the magnetic spectra. The results of these measurements reveal that the (1/2,0) anomaly displays properties more consistent with a spinon-pairs continuum than with a multi-magnon one. We therefore find clear signatures of exotic physics beyond the canonical ferro-orbital, AF Néel ground state, driven by extremely large nearest-neighbour hopping t and possibly next-nearest neighbour hopping t’ [4].

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Email corresponding author: leonardo.martinelli@polimi.it

Prefered presentation – Oral

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Direct estimation of Tc and T* in YBCO by means of RIXS: a new approach to the two gaps phenomenology

Giacomo Merzoni[1,2], Leonardo Martinelli[1], Riccardo Arpaia[3], Nicholas B. Brookes[4], Lucio Braicovich[1,4], Marco Moretti Sala[1], Giacomo Ghiringhelli[1,5]

[1]Dipartimento di Fisica, Politecnico di Milano, piazza Leonardo da Vinci 32, I-20133 Milan, Italy. [2]European XFEL, Holzkoppel 4, 22869, Schenefeld, Germany. [3]Quantum Device Physics Laboratory, Department of Microtechnology and Nanoscience, Chalmers University of Technology, SE-41296 Göteborg, Sweden. [4]ESRF, The European Synchrotron, 71 Avenue des Martyrs, CS 40220, F-38043 Grenoble, France. [5]CNR-SPIN, Dipartimento di Fisica, Politecnico di Milano, I-20133 Milan, Italy

Abstract

The pseudogap phase represents the most inexplicable and peculiar region in the phase diagram of hole doped cuprates [1]. The representation of the pseudogap as a competing phase to superconductivity or rather as a precursor of the superconducting condensate formation is still deeply debated [2]. Although angle resolved photoemission spectroscopy (ARPES) has been providing key information on this phase of matter [3], it is inapplicable to many cuprate families like the $ReBa_2Cu_3O_{7-\delta}$. Other techniques, like transport measurements, have been deeply exploited to study the transition from the pseudogap to the so called ‘strange metal’ phase, offering nonetheless a very indirect estimation of the pseudogap transition temperature value *T**. The lack of a direct assessment of the pseudogap phenomenology makes the overall picture heavily incomplete and therefore the rising of alternative spectroscopic techniques can enrich our understanding of this puzzling regime.

Thanks to the recent improvements in the experimental resolution, resonant inelastic x-ray scattering (RIXS) has been demonstrated to be a valuable technique to study the superconducting gap [4] and a promising candidate to investigate also the pseudogap phase.

Complementing the experimental activity with simulations of the charge dynamical structure factor to find the optimum **q** position to maximize the effects of the pseudogap, we have recently completed a successful experiment at the ID32 beamline (ESRF) [5]. We carried out RIXS measurements on an underdoped $YBa_2Cu_3O_{7-\delta}$ (YBCO) sample ($p \approx 0.15$), where the putative T^* is indirectly estimated to be far from T_c. Performing a high resolution (≈ 40 meV) T dependence at a fixed **q**, we have been able to observe two clear jumps in the integrated low energy spectral intensity, below 100 meV in energy loss. The first change of slope is at T ≈ 87 K, in good agreement with the T_c estimated from transport measurements, the second one at T ≈ 160 K, compatible with T^*. These results clearly demonstrate the possibility to extract both T_c and T^* from RIXS measurements, opening the route to an alternative technique to study the superconducting gap opening behaviour and to assess the nature of the pseudogap to the strange metal transition in a vast range of compounds.

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Time-resolved resonant x-ray scattering in antiferromagnetic quantum materials

Understanding of the transient behavior of magnetic systems is of high interest due to the fundamental physics of emergent quantum states and potential magnetism-based next-generation electronics. In this talk we will discuss recent work on sub-picosecond resolution pump-probe resonant x-ray measurements that revealed a striking contrast between the transient behavior of strongly gapped and nearly gapless prototype antiferromagnetic systems. Discussion of the experimental setup, data acquisition, and data analysis will be emphasized due to the extreme difficulty of such measurements. Finally, the future outlook in light of recent developments at several x-ray free electron laser sources will be discussed. Such advances are expected to make similar experiments much more feasible and user-friendly in the near future.

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Nature of charge density waves in kagome metals

Hu Miao[1], Ayman Said[2]
[1]Oak Ridge National Lab, Oak Ridge, USA. [2]Argonne National Lab, Lemont, USA

Abstract

The intricate interplay between novel lattice geometry and spontaneous symmetry-breaking states is at the forefront of contemporary research on quantum materials. Recently, evidence of unconventional charge density waves are observed in kagome metals where multiple van Hove singularities are near the Fermi level. Here, we use meV-resolution inelastic x-ray scattering to unravel the nature of charge density waves and their interplay with magnetism and superconductivity.

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Ultrafast manipulation of electronic interactions in quantum materials

Denitsa Baykusheva[1], Hoyoung Jang[2], Ali Husain[3], Sangjun Lee[4], Sophia TenHuisen[5], Preston Zhou[1], Sunwook Park[6], Hoon Kim[6], Jin-Kwang Kim[6], Hyeong-Do Kim[2], Minseok Kim[2], Sang-Youn Park[2], Peter Abbamonte[4], Bumjoon Kim[6], Genda Gu[7], Yao Wang[8], Matteo Mitrano[1]

[1]Harvard University, Cambridge, USA. [2]Pohang Accelerator Laboratory, Pohang, Korea, Republic of. [3]University of British Columbia, Vancouver, Canada. [4]University of Illinois at Urbana-Champaign, Urbana, USA. [5]Hav, Cambridge, USA. [6]Pohang University of Science and Technology, Pohang, Korea, Republic of. [7]Brookhaven National Laboratory, Upton, USA. [8]Clemson University, Clemson, USA

Abstract

Intense ultrashort electromagnetic fields are an increasingly important tool to realize and control novel emergent phases in quantum materials. Among a variety of nonthermal excitation pathways, a particularly intriguing route is represented by the direct light-engineering of effective many-body interactions, such as electron hopping amplitudes and electron-electron repulsion. Achieving a light-induced dynamical renormalization of the screened onsite Coulomb repulsion (“Hubbard U”) would have far-reaching implications for high-harmonic generation, attosecond spectroscopy and ultrafast magnetism in the solid state. However, experimental evidence for a dynamically controlled Hubbard U remains scarce. In this talk, I will present a recent demonstration of light-induced renormalization of the Hubbard U in a high-temperature superconductor, $La_{2-x}Ba_xCuO_4$, and discuss its implications for the control of superconductivity and magnetism, as well as for time-

resolved RIXS experiments on light-driven quantum materials.

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New Ultrahigh Resolution 2D-RIXS Facility in Tohoku, Japan

Jun Miyawaki, Kentaro Fujii, Takashi Imazono, Koji Horiba, Yoshiyuki Ohtsubo, Nobuhito Inami, Takeshi Nakatani, Kento Inaba, Akane Agui, Hiroaki Kimura, Masamitu Takahasi
National Institutes for Quantum Science and Technology, Miyagi, Japan

Abstract

A new 3 GeV ring is under construction in Tohoku, Japan and is scheduled to be operational in 2024. In this synchrotron radiation facility, we are developing an ultrahigh resolution 2D-RIXS spectrometer with a dedicated beamline. An APPLE-II type undulator is employed to provide variable polarization (liner horizontal and vertical, and left and right circular polarization) and covers the energy range of 250–2000 eV. The beamline was designed specifically for the 2D-RIXS spectrometer [1–3], and the dispersed light from the beamline monochromator will be irradiated directly onto a sample. The target total energy resolution (combined resolution of the beamline and RIXS spectrometer) is <10meV below $h\nu$=1000 eV. In such ultrahigh resolution, measurement efficiency is one of the factors that determine the energy resolution [4]. Therefore, to improve the lower efficiency at the higher resolution, we employ the 2D-RIXS spectrometer to compensate for the low throughput and realize the RIXS facility with single meV resolution.

We will construct a ~76 m long beamline with ~12 m long RIXS spectrometer which can rotate continuously by ~120° in the longest area in the synchrotron radiation facility. In the 2D-RIXS spectrometer, vertically energy-dispersed X-ray from a beamline monochromator is horizontally focused and irradiated on the sample. An imaging mirror in the 2D-RIXS spectrometer images scattered X-ray from the sample on a position-sensitive detector with keeping its position corresponding to the incident energy. Simultaneously, in the perpendicular direction, the grating in the 2D-RIXS spectrometer

disperses and focuses the scattered X-rays on the detector. Thus, the energies of the incident and scattered X-rays are two-dimensionally resolved on the detector, resulting in high efficiency. The expected resolution and photon flux with a virtual 2 μm wide exit slit are ~5.4 meV at FWHM at 1000 eV and ~1×10^{11} photons/s at 500–1000 eV. Since the field of view of the 2D-RIXS spectrometer is ~120 μm, the photon flux of ~6×10^{12} photons/s is available, and the total throughput is expected to be about 10 times higher than that for a conventional spectrometer that uses monochromatic X-ray. In the presentation, we will introduce the optical design of the beamline and RIXS spectrometer, and outline the improvement of the efficiency and the feasibility of the ultrahigh resolution by the 2D-RIXS spectrometer.

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Quadrupolar magnetic excitations in an isotropic spin-1 antiferromagnet

Abhishek Nag[1], Alberto Nocera[2,3], Stefano Agrestini[4], Mirian Garcia-Fernandez[4], Andrew Walters[4], Sang-Wook Cheong[5], Steven Johnston[6,7], Ke-Jin Zhou[4]

[1]Paul Scherrer Institute, Villigen, Switzerland. [2]Stewart Blusson Quantum Matter Institute, Vancouver, Canada. [3]University of British Columbia, Vancouver, Canada. [4]Diamond Light Source, Didcot, United Kingdom. [5]Rutgers Center for Emergent Materials, Piscataway, USA. [6]The University of Tennessee, Knoxville, USA. [7]Institute for Advanced Materials and Manufacturing, Knoxville, USA

Abstract

The microscopic origins of emergent behaviours in condensed matter systems are encoded in their excitations. In ordinary magnetic materials, single spin-flips give rise to collective dipolar magnetic excitations called magnons. Likewise, multiple spin-flips can give rise to multipolar magnetic excitations in magnetic materials with spin S>=1. Unfortunately, since most experimental probes are governed by dipolar selection rules, collective multipolar excitations have generally remained elusive. For instance, only dipolar magnetic excitations have been observed in isotropic S=1 Haldane spin systems. I will present our Ni L_3-edge resonant inelastic x-ray scattering results on antiferromagnetic S=1 Haldane chain material Y_2BaNiO_5, where we observe a hidden quadrupolar constituent of the spin dynamics. Our results demonstrate that pure quadrupolar magnetic excitations can be probed without direct interactions with dipolar excitations or anisotropic perturbations. Originating from on-site double spin-flip processes, the quadrupolar magnetic excitations in Y_2BaNiO_5 show a remarkable dual nature of collective dispersion. While one component propagates as non-interacting entities, the

other behaves as a bound quadrupolar magnetic wave. This result highlights the rich and largely unexplored physics of higher-order magnetic excitations.

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Doping dependent study of charge ordering phenomena in $YBa_2Cu_3O_{6+x}$ under uniaxial stress

Suguru Nakata[1], Davide Betto[2], Enrico Schierle[3], Hun-ho Kim[1], Sofia M. Souliou[4], Sajna Hameed[1], Yiran Liu[1], Tom L. Lacmann[4], Katrin Fürsich[1], Toshinao Loew[1], Roberto Sant[2], Kurt Kummer[2], Nicholas B. Brookes[2], Eugen Weschke[3], Andrew P. Mackenzie[5], Clifford W. Hicks[5], Bernhard Keimer[1], Matthieu Le Tacon[4], Matteo Minola[1]
[1]Max Planck Institute for Solid State Research, Stuttgart, Germany. [2]European Synchrotron Radiation Facility (ESRF), Grenoble, France. [3]Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany. [4]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, Karlsruhe, Germany. [5]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany

Abstract

Many-body ground states in correlated materials are nearly degenerate due to strong electron correlations and thus quite sensitive to external fields. This is specifically evidenced in layered cuprates where antiferromagnetism, charge-density waves (CDW), and superconductivity can be tuned by doping, magnetic field, and external pressure. Recent high-resolution nonresonant inelastic x-ray scattering (IXS) studies of the high-temperature superconductor $YBa_2Cu_3O_{6.67}$ (doping level $p = 0.12$ holes per Cu ion) have revealed that uniaxial pressure induces a substantial enhancement of the two-dimensional (2D) CDW elastic scattering compared to previous works without strain [1]. Crucially, we discovered that uniaxial pressures large enough, typically around 1% of compression along the a-axis of $YBa_2Cu_3O_{6+x}$, induce a three-dimensional (3D) long-range-ordered CDW state along the b-axis, akin to that observed in the presence of high magnetic field but with correlation

volumes and up to temperatures exceeding those obtained using field. The IXS measurements also showed that the strong softening of a phonon mode drives the formation of the 3D-CDW. More recently, we have also carried out Cu-L_3 resonant inelastic x-ray scattering (RIXS) experiments at the ESRF, confirming the enhancement of the 2D-CDW always along the direction perpendicular to that of the applied uniaxial pressure [2]. The results provide an exciting new perspective to control competing phases in cuprates, but the limited counting rate of energy-resolved experiments prevented us from mapping the strain- and temperature-dependence of both 2D- and 3D-CDW. For this reason, only one particular doping level $p = 0.12$ (1/8 doping) where the CDW is the strongest has been investigated so far.

Here, I will report resonant energy-integrated x-ray scattering (REXS) measurements of $YBa_2Cu_3O_{6+x}$ at various doping levels to obtain a systematic doping dependence of the uniaxial stress effects on CDW in $YBa_2Cu_3O_{6+x}$. The 3D CDW was observed only in the vicinity of 1/8 doping and shows a strong competition with superconductivity, while the 2D CDW was enhanced in a wider doping regime. Moreover, I will present new Cu-L_3 RIXS data where the uniaxial pressure was applied already at high temperature before cooling that show a partial suppression of the CDW with the modulation parallel to the pressure direction, thus shining light on the mechanism of condensation of charge density fluctuations in static CDW. Our comprehensive observations yield new insight into anomalous normal-state properties of cuprates and demonstrate the complementarity of energy-resolved and energy-integrated x-ray scattering experiments to study uniaxial stress effects of competing phases in quantum materials.

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Signature of Many-Body Localization of Phonons in Strongly Disordered Superlattices

Thanh Nguyen[1], Nina Andrejevic[1], Hoi Chun Po[1], Qichen Song[1], Yoichiro Tsurimaki[1], Nathan Drucker[2], Ahmet Alatas[3], Esen Alp[3], Bogdan Leu[3,4], Alessandro Cunsolo[5], Yong Cai[6], Lijun Wu[6], Joseph Garlow[6], Yimei Zhu[6], Hong Lu[7], Arthur Gossard[8], Alexander Puretzky[9], David Geohegan[9], Shengxi Huang[10], Mingda Li[1]
[1]Massachusetts Institute of Technology, Cambridge, USA. [2]Harvard University, Cambridge, USA. [3]Argonne National Laboratory, Lemont, USA. [4]Miami University, Oxford, USA. [5]University of Wisconsin at Madison, Madison, USA. [6]Brookhaven National Laboratory, Upton, USA. [7]Nanjing University, Nanjing, China. [8]University of California, Santa Barbara, Santa Barbara, USA. [9]Oak Ridge National Laboratory, Oak Ridge, USA. [10]The Pennsylvania State University, University Park, USA

Abstract

The experimental realization of many-body localization (MBL) in solid-state systems has remained challenging. In parallel, open questions remain on the localization of phonons as the strong interactions and disorders in phononic superlattice systems would call for an MBL description. The elucidation of phonon localization through the lens of MBL would have major implications for connecting the MBL phenomenology to a mesoscopic solid-state system. In this talk, I will report evidence of a possible phonon MBL phase in disordered GaAs/AlAs superlattices. Using grazing-incidence inelastic X-ray scattering, we observe a strong deviation of the phonon population in samples doped with ErAs nanodots at low temperature, signaling a departure from thermalization. This behavior occurs within finite phonon energy and wavevector windows, suggesting a localization-thermalization crossover. We

support our observation by proposing a theoretical model for the effective phonon Hamiltonian in disordered superlattices, and showing that it can be mapped exactly to a disordered 1D Bose–Hubbard model with a known MBL phase. Our study opens up new opportunities as a new experimental solid-state platform for realizing possible MBL phenomena, and as a novel phonon phase far away from equilibrium.

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Probing Spin Excitations in an Ultra-Thin van der Waals Helimagnet

Connor Occhialini[1], Qian Song[1], Yi Tseng[1], Joshua Sanchez[1], Luiz Martins[1], Valentina Bisogni[2], Jonathan Pelliciari[2], Riccardo Comin[1]
[1]Department of Physics, Massachusetts Institute of Technology, Cambridge, MA, USA. [2]National Synchrotron Light Source II, Brookhaven National Laboratory, Upton, NY, USA

Abstract

Materials with non-collinear magnetic structures host many unique phenomena, including multiferroic, magnetoelectric and topological phases [1]. Among these, the discovery of the persistence of non-collinear magnetism in the van der Waals (vdW) nickel dihalides (NiX_2, X = Br, I) down to the few- and single-atomic-layer limit paves the way for the realization of these properties in 2D devices/heterostructures [2-4]. The geometric frustration of the triangular lattice in these materials leads to a complex magnetic phase diagram where the ground state is determined by a delicate balance of intra-layer exchange interactions [5]. However, from an experimental perspective, accessing these interactions in the 2D limit is challenging due to the limited thickness. In this work, we address these challenges and present measurements of the dispersive magnetic excitations in NiI_2 in the bulk and ultra-thin limit using resonant inelastic X-ray scattering (RIXS) [6]. We uncover the dispersion of a single-magnon branch in the bulk which shows a clear correspondence to the incommensurate helimagnetic order [6,7]. Additional measurements performed on an ultra-thin sample reveal dispersive spin excitations with marked renormalization with respect to the bulk. Based on these results, we assess the evolution of the intra-layer exchange interactions and the helimagnetic order in the 2D limit of NiI_2. More broadly, these measurements provide information regarding the nature of finite-momentum spin excitations in layered 2D magnets, which have thus far eluded direct observation, and establish RIXS as an ideal technique to investigate

complex magnetic phases in vdW materials.

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Unimolecular decomposition of high explosives observed by X-ray Raman scattering spectroscopy

Oscar Paredes Mellone
1 SLAC Stanford Linear Accelerator, Menlo Park, USA. 2 Lawrence Livermore National Laboratory, Livermore, USA

Abstract

Oscar A. Paredes Mellone1, Dimosthenis Sokaras1, Trevor Willey2, Michael Nielsen2, Arianna Gleason-Holbrook1.

1 SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, California 94025 USA.

2 Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, California 94551 USA.

Abstract

Theory and modeling have long preceded experiment in the fundamental physical and chemical kinetic properties of detonation. This technical gap exists because conventional chemical analytic methods are not easily applied to the rapid processes within the tumultuous dense bulk of the detonation. Hard X-ray core-level spectroscopies give access to local electronic structure within the bulk and are sensitive to the bond nature of the exciting atom. X-ray Raman spectroscopy (XRS), a core-level photon-in/photon-out hard X-ray technique, emerges as a unique tool to address these fundamental questions. XRS provides bulk sensitivity and allows investigating absorption edges of low Z elements with hard X-rays avoiding usual constraints inherent to UV/soft X-ray spectroscopies.

We present X-ray Raman spectra of high explosives measured at the BL-15 beamline of the Stanford Synchrotron Radiation Facility1, along with first-principle density functional theory calculations. By combining experimental results and theoretical calculations, spectral features and trends observed in the near-edge region of the XRS spectra for different radiation dose were related to unimolecular decomposition intermediates proposed in the literature2-5. By identifying these spectral features in undetonated explosives, investigating the achievable signal-to-noise intensity with X-ray flux, and overall addressing the sensitivity of the technique to unravel the kinetics of high explosive chemical decomposition, this work paves the way for dynamic X-ray Raman scattering spectroscopy in future in-situ detonation time-resolved experiments.

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Email corresponding author: oparedes@slac.stanford.edu

Preferred presentation - Oral

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Correlations and Entanglement in van der Waals Antiferromagnets

Je-Geun Park
Seoul National University, Seoul, Korea, Republic of

Abstract

Two-dimension (2d) is most interesting from the physics viewpoint because of the generic dimensional fluctuations. And there are three fundamental Hamiltonians of magnetism: Ising, XY, and Heisenberg models. The new class of van der Waals (vdW) antiferromagnets discovered in the mid-2010s [1,2] has since been proven an ideal playground to explore the three spin models [3-5].

In addition, they also offer possibilities for the study of correlation and entanglement on 2d. True, both concepts on their own have been at the center of intense attention and activities. However, 2d systems harboring these two interesting physics will be a huge advantage. We have been exploring several vdW antiferromagnets with that particular motivation. I will take $NiPS_3$ [6,7] and NiI_2 [8,9] for the question of entanglement in 2d materials.

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Evolution from charge-order phase to high-temperature superconductivity

Qizhi Li[1], Changwei Zou[1], Jaewon Choi[2], Mirian Garcia-Fernandez[2], Ke-Jin Zhou[2], Hsiao-Yu Huang[3], Di-Jing Huang[3], Eugen Weschke[4], Yayu Wang[5], Yingying Peng[1]

[1]Peking University, Beijing, China. [2]Diamond Light Source, Didcot, United Kingdom. [3]National Synchrotron Radiation Research Center, Hsinchu, Taiwan. [4]Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany. [5]Tsinghua University, Beijing, China

Abstract

The origin of high-Tc superconductivity (HTSC) of copper oxide superconductors remains one of the most important challenges in condensed matter physics. HTSC can be obtained through doping a Mott insulator or cooling a strange metal, and great efforts have been put forward to understand how Cooper pairs form and condense in the past 36 years. However, there is still no consensus on the critical process to the emergence of superconductivity when electron-electron correlations dominate. Recently we have carried out high-resolution resonant inelastic X-ray scattering and scanning tunneling microscopy studies, combining bulk and surface, momentum- and real-space information to address this issue. In this talk, I will present our recent results on Bi2212 [1] and LSCO [2] cuprates. These include: (i) How high-temperature superconductor emerges near the onset of Tc dome [1]; (ii) How charge excitations and phonons evolve as a function of doping and temperature [2]. Our results suggest that high-temperature superconductivity emerges out of the charge-ordered phase.

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Evolution from charge-order phase to high-temperature superconductivity

Yingying Peng[1], Changwei Zou[1], Jaewon Choi[2], Qizhi Li[1], Riccardo Arpaia[3], Leonardo Martinelli[3], Chaohui Yin[4], Mirian Garcia-Fernandez[2], Stefano Agrestini[2], Xingjiang Zhou[4], Giacomo Ghiringhelli[3], Nicholas Brookes[5], Ke-Jin Zhou[2], Yayu Wang[6]
[1]Peking University, Beijing, China. [2]Diamond Light Source, Didcot, United Kingdom. [3]Politecnico di Milano, Milan, Italy. [4]Institute of Physics, Beijing, China. [5]The European Synchrotron, Grenoble, France. [6]Tsinghua University, Beijing, China

Abstract

The origin of high-Tc superconductivity (HTSC) of copper oxide superconductors remains one of the most important challenges in condensed matter physics. HTSC can be obtained through doping a Mott insulator or cooling a strange metal, and great efforts have been put forward to understand how Cooper pairs form and condense in the past 36 years. However, there is still no consensus on the critical process to the emergence of superconductivity when electron-electron correlations dominate. Recently we have carried out high-resolution resonant inelastic X-ray scattering and scanning tunneling microscopy studies, combining bulk and surface, momentum- and real-space information to address this issue. In this talk, I will present our recent results on bi-layer Bi2212 and tri-layer Bi2223 cuprates. These include: (i) How high-temperature superconductor emerges near the onset of Tc dome [1]; (ii) How charge excitations and phonons evolve as a function of doping and temperature in the presence of robust Tc of 110 K in overdoped Bi2223 [2]. Our results suggest that the Cooper pairs grow out of a charge-ordered state, and then condense accompanied by an enhanced interplay between charge excitations and electron-phonon coupling.

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Email corresponding author: yingying.peng@pku.edu.cn

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Ultrafast dynamics in quantum matter at SwissFEL: capabilities of Furka endstation.

Elia Razzoli, Eugenio Paris, Cristian Svetina, Hiroki Ueda, Biaolong Lui, Luc Patthey
Paul Scherrer Institut, Villigen, Switzerland., Villigen, Switzerland

Abstract

Time-resolved spectroscopies have provided insights in the quest for understanding the fundamental properties of quantum materials and towards controlling their functional properties through light-matter interaction. In this regard, Free Electrons Lasers (FEL) have developed as a powerful tool to perform ultrafast x-ray spectroscopy allowing to obtain energy and momentum-resolved information.

In this contribution, I will introduce the SwissFEL soft-X-ray condensed matter experimental endstation, named Furka, which is dedicated to time-resolved X-ray absorption (TR-XAS), resonant X-Ray diffraction (TR-RXRD) and Resonant Inelastic X-ray Scattering (TR-RIXS) experiments to study quantum materials. The current status of the endstation and the foreseen timeline for its commissioning and user operation will also be discussed.

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Nematic Correlation Length in Iron-Based Superconductors Probed by Inelastic X-Ray and Neutron Scattering

Dmitry Reznik[1], Adrian Merritt[1], Frank Weber[2], John-Paul Castellan[2,3], Thomas Wolf[2], Daisuke Ishikawa[4], Ayman Said[5], Ahmet Alatas[6], Rafael Fernandes[7], Alfred Baron[4]
[1]University of Colorado-Boulder, Boulder, CO, USA. [2]Karlsruhe Institute of Technology, Karlsruhe, Germany. [3]Laboratoire Leon Brillouin, Saclay, France. [4]RIKEN SPring-8 Center, Hyogo, Japan. [5]Argonne National Lan, Lemont, IL, USA. [6]Argonne National Lab, Lemont, IL, USA. [7]University of Minnesota, Mineapolis, MN, USA

Abstract

Nematicity is ubiquitous in electronic phases of high-Tc superconductors, particularly in the Fe-based systems. We used inelastic x-ray scattering to extract the temperature dependent nematic correlation length ξ from the anomalous softening of acoustic phonon modes in FeSe, underdoped $Ba(Fe_{0.97}Co_{0.03})_2As_2$, and optimally doped $Ba(Fe_{0.94}Co_{0.06})_2As_2$. In all cases, we find that ξ is well described by a power law $(T-T_0)^{-1/2}$ extending over a wide temperature range. Combined with the previously reported Curie-Weiss behavior of the nematic susceptibility, these results point to the mean-field character of the nematic transition, which we attribute to a sizable nematoelastic coupling that is likely detrimental to superconductivity. New inelastic neutron scattering results on phonon linewidths will also be discussed.

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Electronic phase transitions in quantum 3d materials at high pressure by RIXS : the case of $SrFeO_2$ and $BaFe_2Se_3$

Jean-Pascal Rueff[1,2], Victor Balédent[3,4]
[1]Synchrotron SOLEIL, Gif-sur-Yvette, France. [2]Sorbonne Université, Paris, France. [3]Laboratoire de Physique des Solides, Orsay, France. [4]Université Paris Saclay, Gif-sur-Yvette, France

Abstract

Pressure is an effective mean to alter the structural, electronic and magnetic response of 3d quantum materials as the change of the atomic distances affect the electron density, orbital hybridization and crystal electric field strength. RIXS appears as a powerful tool of characterization of materials under high pressure [1]. Here we will illustrate this approach through recent RIXS high pressure studies realized at GALAXIES beamline, SOLEIL synchrotron. We will address more specifically the stability of the spin state in the square planar Fe compound $SrFeO_2$ [2] and the interplay with superconductivity in the $BaFe_2Se_3$ spin ladder compounds [3]. Perspectives for high pressure RIXS studies at GALAXIES beamline in line with the Upgrade of SOLEIL will be discussed.

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An Overview of X-ray Compton Scattering: Basic and Applied Research

Yoshiharu Sakurai
Japan Synchrotron Radiation Research Institute (JASRI), Sayo, Japan

Abstract

An overview of recent works in the field of X-ray Compton scattering is presented. The X-ray Compton scattering technique measures the electron momentum density distribution, and thus provides information on Fermi surfaces and electron orbitals. Recent studies include the nature of Fermi surfaces in high-Tc cuprates [1] and Kantor-Wu disordered alloys [2], and the orbital characters of lithium-ion battery materials [3]. A new direction in this technique is Compton scattering imaging (CSI), in which the material behaviors inside palm-scale products, such as lithium-ion batteries, are visualized [4]. Recent works of CSI include the visualization of liquid water in fuel cell materials [5]. In this talk, material behaviors in fuel cells are also presented.

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High resolution RIXS study of in SrTiO3 and SrTiO3-Based Heterostructures

marco salluzzo[1], Lucio Braicovich[2,3], Nicholas B. Brookes[2], Giacomo Ghiringhelli[3], YyngYing Peng[4], Keith Gilmore[5], Andrey Geondzhian[6]
[1]CNR-SPIN, Napoli, Italy. [2]ESRF, Grenoble, France. [3]Dipartimento di Fisica Politecnico di milano, Milano, Italy. [4]International Center for Quantum Materials,School of Physics, Peking University, Beijing, China. [5]Institute for Physics Humboldt University of Berlin, Berlin, Germany. [6]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

Abstract

Quasiparticles in metals and semiconductors are formed through the interactions of electrons (holes) with the elementary excitations of the solid, like phonons and magnons. Here, by using titanium L-edge resonant inelastic x-ray scattering (RIXS) with unprecedent energy resolution we demonstrate that the quasiparticles in bulk SrTiO3 and related heterostructures are large polarons formed by the coupling of itinerant carriers to optical phonons. RIXS spectra on different samples, from barely undoped SrTiO3 to strongly metallic heterostructures, show low (25-30 meV), mid (50-60 meV) and high energy (90-100 meV, LO3) optical phonons excitations and, at a higher energy (125-135 meV), an intra-t2g d-d excitation accompanied by the emission of an LO3 optical phonon, which represents a hallmark of large polarons in STO and LAO/STO bilayers. Furthermore, from the analysis of the RIXS cross section, we find that the electron-phonon coupling constant of the LO3 phonon mode decreases with the carrier density as consequence of the screening of the large-polaron self-induced polarization [1].

Beside confirming earlier signatures by ARPES at the surface of STO [2], in LAO/STO [3] and in FeSe/STO bilayers [4], our study demonstrates more generally the emergence of large polaron

physics in both bi- and three-dimensional titanates. Finally, it emerges that polarons are observed also in nominally undoped STO, with a coupling constant well below the value expected for small polarons formation. Consequently, we can infer that even at the very low doping level, as that induced by residual defects or by long living photodoped carriers, 3d1 electrons are dressed by long-range polar lattice distortions, as theoretically predicted in other wide band gap materials like LiF [5]. Future investigations and theoretical modeling of the normal and superconducting state of STO and STO-based heterostructures will have to take in consideration the central role of large polarons in these materials

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Theoretical investigation on Kondo physics of Ce intermetallicsby means of 3*d*-4*f* Resonant Inelastic X-ray Scattering

Norimasa Sasabe[1], Takayuki Uozumi[2]

[1]Japan Synchrotron Radiation Research Institute, SPring-8 Kouto, Sayo, Hyogo, Japan. [2]Graduate School of Engineering, Osaka Metropolitan University, Sakai, Osaka, Japan

Abstract

The various phenomena of Ce intermetallics have much attracted; Kondo effects, Multipole ordering, Non-Fermi liquid behavior, and unconventional superconductivity are realized owing to the Ce 4*f* electron character [1-3]. For a long term, a large number of investigations on Ce intermetallics have been performed, and, especially, the studies using core-level spectroscopies have contributed to an estimation of valence number and solid state parameters. In order to clarify the mechanism of the various phenomena, these acquirements from core-level spectroscopies have been linked together with the direct physical quantities, such as Kondo temperature.

In this conference, we show the spectral calculations of 3*d*-4*f* resonant inelastic X-ray scattering (RIXS) for the electronic state of $AuCu_3$-type Ce intermetallics, $CeIn_3$ and $CeSn_3$, and discuss that the RIXS measurement can directly detect the binding energy of Kondo singlet, which is related to the Kondo temperature. Furthermore, we show that an itinerant component arising from the *f*-*c* hybridization of a singlet formation can be observed by setting appropriately the polarization and energy of incident x-ray photons under 3*d*-4*f* RIXS measurement [4]. In addition to the binding energy, the itinerant component is also important to understand the 4*f* electron state of Ce intermetallics which is accompanied by an appearance of the various phenomena.

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Time-resolved RIXS at SCS instrument, European XFEL – design and commissioning of the Heisenberg RIXS User Consortium spectrometer

Justine Schlappa[1], Benjamin van Kuiken[1], Zhong Yin[1], Sergii Parchenko[1], Martin Teichmann[1], Natalia Gerasimova[1], Piter Miedema[1], Giuseppe Mercurio[1], Robert Carley[1], Carsten Broers[1], Jan Torben Delitz[1], Luigi Adriano[1], Manuel Izquierdo[1], Andreas Scherz[1], Yingying Peng[2], Giacomo Claudio Ghiringhelli[2], Stefan Neppl[3,4], Friedmar Senf[3,4], Frank Siewert[3], Christian Weniger[3], Annette Pietzsch[3], Alexander Foehlisch[3,4], Simo Huotari[5], Torben Reuss[6], Simone Techert[6], Tim Laarmann[6]
[1]European XFEL, Schenefeld, Germany. [2]Politecnico di Milano, Milano, Italy. [3]HZB, Berlin, Germany. [4]University of Potsdam, Potsdam, Germany. [5]University of Helsinki, Helsinki, Finland. [6]DESY, Hamburg, Germany

Abstract

The User Consortium Heisenberg RIXS (hRIXS) spectrometer was built in order to enable soft x-ray time-resolved resonant inelastic x-ray scattering (tr-RIXS) studies at the European X-ray Free Laser Facility (European XFEL). The design goal of the spectrometer was on one hand a high energy resolution, potentially allowing the exploration of the Heisenberg limit for tr-RIXS and on the other hand a possibly effortlessness operation. The scientific motivation of hRIXS was enabling tr-RIXS studies for the two large user communities, one with focus on complex and quantum materials and the other one with focus on chemical systems (including a liquid-jet environment).

The hRIXS spectrometer was installed at the Spectroscopy and Coherent Scattering (SCS) instrument [1,2], in 2020/2021. The technical and x-ray commissioning took place in May 2021. Very recently, first tr-RIXS commissioning experiments took place, in close collaboration with the user communities. The results look very promising and indicate that hRIXS, in combination with the unique features of the European XFEL and the SCS beamline, provides currently the best performance for soft x-ray RIXS studies at an FEL.

In my talk I will present the design of the hRIXS spectrometer and give an overview of the available parameters. Further, I will present some of the commissioning results and discuss the currently obtained performance. I will conclude with an outlook for tr-RIXS studies with hRIXS at the SCS instrument.

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Spin-excitation anisotropy in the nematic state of detwinned FeSe

Thorsten Schmitt[1], Xingye Lu[2]

[1]Paul Scherrer Institut, Photon Science Division, CH-5232, Villigen PSI, Switzerland. [2]Center for Advanced Quantum Studies and Department of Physics, Beijing Normal University, Beijing 100875, China

Abstract

The interplay among magnetism, electronic nematicity, and superconductivity is a key issue in strongly correlated materials including iron-based, cuprate, and heavy-fermion superconductors. Magnetic fluctuations have been widely discussed as a pairing mechanism of unconventional superconductivity, but recent theory predicts that quantum fluctuations of electronic nematicity, which is characterized by rotational symmetry breaking, may also promote high-temperature superconductivity.

Iron selenide (FeSe) is a unique material among iron-based superconductors because of its simple crystal structure consisting of stacked charge-neutral FeSe layers and unusual electronic properties, such as the strong anisotropy of the superconducting order parameter, an extended electronic nematic phase, and highly tunable T_C. In particular, different from iron pnictides, which have a collinear antiferromagnetic (AF) ground state below the tetragonal-to-orthorhombic structural (nematic) phase transition T_S, FeSe exhibits a similar nematic ransition (T_S ~ 90 K), but has no static AF order. The origin of the electronic nematicity in FeSe is one of the most important unresolved puzzles in the study of iron-based superconductors. In both spin- and orbital-nematic models, the intrinsic magnetic excitations at $\mathbf{Q_1}$ = (1, 0) and $\mathbf{Q_2}$ = (0, 1) of twin-free FeSe are expected to provide decisive criteria for clarifying this issue. Although a spin-fluctuation anisotropy below 10 meV between

Q_1 and **Q_2** has been observed by inelastic neutron scattering around T_c ~ 9 K (<< T_s ~ 90 K), it remains unclear whether such an anisotropy also persists at higher energies and associates with the nematic transition T_s.

Here we use resonant inelastic x-ray scattering at the Fe L_3-edge to probe the high-energy magnetic excitations of uniaxial-strain detwinned FeSe and $BaFe_2As_2$ [1]. A prominent anisotropy between the magnetic excitations along the H and K directions is found to persist to ~ 200 meV in FeSe, which is even more pronounced than the anisotropy of spin waves in $BaFe_2As_2$. This anisotropy decreases gradually with increasing temperature and finally vanishes at a temperature around the nematic transition temperature T_s. Our results reveal an unprecedented strong spin excitation anisotropy with a large energy scale well above the d_{xz}/d_{yz} orbital splitting, suggesting that the nematic phase transition is primarily spin-driven [1]. Moreover, the measured high-energy spin excitations are dispersive and underdamped, which can be understood from a local-moment perspective. Our findings provide the much-needed understanding of the mechanism for the nematicity of FeSe and points to a unified description of the correlation physics across seemingly distinct classes of Fe-based superconductors.

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Soft X-ray imaging spectroscopy with micrometre resolution

Jan Oliver Schunck[1,2], Florian Döring[3], Benedikt Rösner[3], Jens Buck[4,5], Robin Yoel Engel[1,2], Piter Miedema[1], Sanjoy Kr Mahatha[5], Moritz Hoesch[1], Adrian Petraru[6], Hermann Kohlstedt[6], Christian Schüßler-Langeheine[7], Christian David[3], Kai Rossnagel[4,5], Martin Beye[1,2]

[1]Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany. [2]Department of Physics, University Hamburg, Hamburg, Germany. [3]Paul Scherrer Institut, Villigen, Switzerland. [4]Institut für Experimentelle und Angewandte Physik, Kiel University, Kiel, Germany. [5]Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany. [6]Nanoelektronik, Technische Fakultät, Kiel Univeristy, Kiel, Germany. [7]Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany

Abstract

Quantum materials derive their intriguing functionalities from a complex energy landscape, which may cause the spatial coexistence of phases with distinct electronic behaviour. However, the classical form of soft X-ray spectroscopies, like X-ray absorption or resonant inelastic X-ray scattering, average the measured spectrum from the entire illuminated spot on the sample – potentially also averaging across regions with different properties.

This contribution presents first results of an experimental setup for soft X-ray imaging spectroscopy with a current spatial resolution down to 1.8 µm [1]. At the heart of the setup is an off-axis transmission Fresnel zone plate, dispersing X-rays scattered from the sample while also imaging the sample in the non-dispersive direction of the zone plate. The one-dimensional focusing can further be

exploited by opening the monochromator exit slit and focusing X-rays incident on the sample to a line, along which the photon energy varies. This scheme opens various applications, like imaging sample structures with RIXS as a contrast mechanism, the $h\nu^2$-scheme [2, 3] to speed up the acquisition of RIXS maps, but also highly efficient spatio-spectroscopic analyses of absorption spectra of bulk materials or of spatial distributions of (resonant) scattering signals.

With this setup, the insulator-metal transition of two compounds was investigated: Firstly, imaging partial fluorescence yield X-ray absorption on micron-sized vanadium dioxide (VO_2) structures (30 μm^2) reveals a phase transition onset of the structure edges at lower temperatures as compared to the centres of the same structures [4]. Secondly, the temperature-dependent distribution of coexisting metallic and insulating regions in the Ca-doped titanate $Y_{0.63}Ca_{0.37}TiO_3$ [5] was studied with imaging soft X-ray Bragg scattering.

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Full phonon softening above the charge-density-wave phase transition in *2H*-$TaSe_2$

Xingchen Shen, Frank Weber
Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, Karlsruhe, Germany

Abstract

Research on charge-density-wave (CDW) ordered transition-metal dichalcogenides continues to unravel new states of quantum matter correlated to the intertwined lattice and electronic degrees of freedom. Here, we report an inelastic x-ray scattering investigation of the lattice dynamics of the canonical CDW compound *2H*-$TaSe_2$ complemented by angle-resolved photoemission spectroscopy. Our results provide evidence for a previously unknown precursor phase above the CDW transition temperature T_{CDW}. The phase at temperatures between T^* (= 128.7 K) and T_{CDW} (= 121.3) is characterized by a fully softened phonon mode, an increasing static CDW superlattice peak and no detectable change in the electronic band structure on cooling below T^*. Thus, *2H*-$TaSe_2$ exhibits increasing structural before electronic order emphasizing the important lattice contribution to CDW transitions. Furthermore, our results rule out the central-peak scenario for the CDW transition in *2H*-$TaSe_2$.

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Real-space time-domain analysis of liquid dynamics using inelastic X-ray scattering

Yuya Shinohara
Oak Ridge National Laboratory, Oak Ridge, USA

Abstract

We report on the real-space time-domain analysis of liquid dynamics using inelastic X-ray scattering. The conventional approach to studying liquid dynamics is to measure the dynamic structure factor, S(Q, ω), where Q and E are the momentum transfer and energy transfer, by inelastic X-ray or neutron scattering and to fit models in Q-ω space to interpret the data. For the systems which lack periodicity in their atomic structures, such as liquids and glasses, analyses in reciprocal space are sometimes powerless in describing their structure and dynamics. Real-space analyses can provide their local information as has been demonstrated in the structural analyses using the pair distribution function. Recent progress in inelastic X-ray and neutron scattering makes it possible to obtain the dynamic structure factor over a wide range of Q and E with a high Q- and E-resolution within a reasonable amount of time. The wide Q- and E-ranges make it possible to calculate the self-part and the distinct part of the Van Hove function–spatial and temporal correlation function [1]–via Fourier transform of S(Q, ω) over ω and Q [2]. With this approach, we have studied spatial and temporal correlations of molecular motion in liquids [2–4]. In this presentation, our recent efforts in understanding the real-space local motion in time domain using the Van Hove function will be presented.

Work supported by the US Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Science and Engineering Division. The IXS experiments at BL43LXU, SPring-8 were carried out under the approval of RIKEN.

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Electronic excitations via Inelastic X-ray Scattering: Green's functions approach.

Francesco Sottile
LSI, Ecole Polytechnique, Palaiseau, France

Abstract

Excitonic effects have proved to be crucial for the quantitative description of
optical absorption [1]. This picture has been confirmed for core excitations [2].
The description of excitons can be conducted, within the many-body Green's
functions (GF) formalism, via the solution of the Bethe-Salpeter equation (BSE), which takes the electron-hole interaction, in the presence of all the
other electrons, automatically into account. More recently, we have put a lot of
effort into extending the applicability of the BSE towards: i) different spectroscopies (like resonanti, non-resonant and coherent IXS [3-5]), ii) unconventional picture (excitonic band-structure [6]), and iii) pictorial tools
(of the electron density evolution). In this seminar, I will illustrate the achievements and problems of theoretical approaches through the GF in tackling
these new spectroscopies and tools.

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Inelastic X-ray Scattering at the Dynamics Beamline P01 at DESY in Hamburg

Hlynur Gretarsson[1,2,3], Martin Sundermann[1,3], Frank-Uwe Dill[1], Simon Mayer[1], Ilya Sergeev[1]

[1]Deutsches Elektron-Synchrotron DESY, Hamburg, Germany. [2]Max-Planck Institute for Solid State Research, Stuttgart, Germany. [3]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany

Abstract

Two instruments at P01 are dedicated to non-resonant and resonant IXS, respectively, which are operated by Max-Planck-Society, but also open to external users. Both instruments utilize spherical analyzers and Rowland type geometry optimized to fulfill the demands of the different experiments. Here we present the characteristics of these setups and a short overview of possible applications.

The non-resonant IXS (NIXS) instrument enables the bulk-sensitive observation of core-level excitations (X-ray Raman scattering) in the soft x-ray regime with hard x-rays.
This allows the element specific study of the electronic structure in complex sample environments like e.g. under high pressure and in batteries. Another focus is making use of beyond dipole transitions, which gives access to study materials in a manner not accessible when using resonant techniques.

The resonant IXS (RIXS) instrument aims to enable high energy resolution RIXS in the tender x-ray regime utilizing hard x-ray optics. At the Ru L3 edge (2.8keV) an energy resolution of about 75meV has been achieved and the instrument recently became open to users. Dispersion of magnons and orbital excitations in ruthenates can be measured with the RIXS instrument. The spectrometer design is versatile and can be adopted to different atomic resonances in the tender x-ray range.

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First tender x-ray RIXS results at the U M_5 edge

Martin Sundermann[1,2], Andrea Marino[1], Denise Christovam[1], Andrea Amorese[1,3], Ayman Said[4], Bernhard Keimer[5], Hlynur Gretarsson[1,2,5], Liu Hao Tjeng[6], Andrea Severing[3,6]
[1]Max-Planck-Institute for Chemical Physics of Solids, Dresden, Germany. [2]Deutsches Elektron-Synchrotron DESY, Hamburg, Germany. [3]University of Cologne, Institute of Physics II, Cologne, Germany. [4]Argonne National Lab, Lemont, USA. [5]Max-Planck Institute for Solid State Research, Stuttgart, Germany. [6]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany

Abstract

The ground state charge density of the 5*f* electrons in uranium compounds can be identified with x-ray Raman using non-resonant inelastic x-ray scattering (NIXS) [1,2,3] but it remains a challenge to get experimental information about the excited states, especially for intermetallic uranium compounds.

At the example of semiconducting UO_2 will be shown that RIXS experiments at the U N_4-edge in the soft x-ray regime (780eV) show multiplet and crystal-field excitations, in contrast to the N_5-edge that has next to no RIXS intensity [4]. It will further be shown that trials on intermetallic uranium compounds however failed to observe any excitations. Seemingly, U N_4-edge RIXS is limited to the more localized U compounds.

In this context the U M_5 edge at 3.5 keV offers an alternative route as already indicated by XES results of Kvashnina et al. [5]. Here we will show RIXS data at the U M_5 edge of UO_2 but also from intermetallic U compounds using the IRIXS end-station at the P01 beamline with its recently installed high resolution (150meV) quartz

analyzer crystals. At the example of UO_2, we show that far more multiplets have a sizable cross-section at the M_5 edge, i.e. the M_5 edge contains more information than the N_4 edge.

Furthermore, data of UGa_2 and URu_2Si_2 show the power of M_5-edge RIXS for intermetallic U compounds. Hence, the M_5-edge RIXS is the way forward for the investigation of the electronic structure of uranium and possibly also transuranium compounds because not only the cross-sections are favorable, in addition, cleaving in the tender x-ray regime is obsolete.

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Nonmagnetic $J = 0$ State and Spin-Orbit Excitations in K_2RuCl_6

Hiroto Takahashi[1,2], Hakuto Suzuki[1], Joel Bertinshaw[1], Sebastian Bette[1], Claus Mühle[1], Jürgen Nuss[1], Robert Dinnebier[1], Alexander Yaresko[1], Giniyat Khaliullin[1], Hlynur Gretarsson[3], Tomohiro Takayama[1,4], Hidenori Takagi[1,2,4], Bernhard Keimer[1]

[1]Max Planck Institute for Solid State Research, Heisenbergstrasse 1, D-70569 Stuttgart, Germany. [2]Department of Physics, The University of Tokyo, Bunkyo-ku, Tokyo 113-0033, Japan. [3]Deutsches Elektronen-Synchrotron DESY, Notkestrstrasse 85, D-22607 Hamburg, Germany. [4]Institute for Functional Matter and Quantum Technologies, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

Abstract

Experiments have found enigmatic nonmagnetic states in insulating Ru^{4+} compounds, where each ion is expected to carry $S = 1$ spin according to Hund's rule. While the nonmagnetism has been mostly explained by a quantum singlet formation by inter-ionic interaction between the spins accompanied by orbital ordering, antiparallel alignment of spin and orbital momenta driven by intra-ionic spin-orbit coupling (SOC) can also result in nonmagnetic $J = 0$ singlet [1]. The observation of the longitudinal Higgs mode in Ca_2RuO_4 indeed suggests a proximate quantum phase transition into the SOC-driven $J = 0$ singlet state [2]. However, materials issues have precluded the identification of spectroscopic fingerprint of the singlet formation mechanism.

We performed Ru L_3-edge resonant inelastic x-ray scattering measurement in K_2RuCl_6, which retains cubic symmetry [3]. Van-Vleck-like small magnetic susceptibility of K_2RuCl_6 down to low

temperature suggests the formation of a nonmagnetic singlet state. Observed RIXS spectrum (Fig. 1) demonstrates excitations from the $J = 0$ ground state to $J = 1$, 2 states (red lines), whose energy levels agree with the numerical calculations within the LS-coupling scheme. Our result confirms the $J = 0$ singlet state as a generic starting point of magnetism in insulating Ru^{4+} compounds, and encourages research into the quantum critical point which lies between the nonmagnetic K_2RuCl_6 and magnetic Ca_2RuO_4.

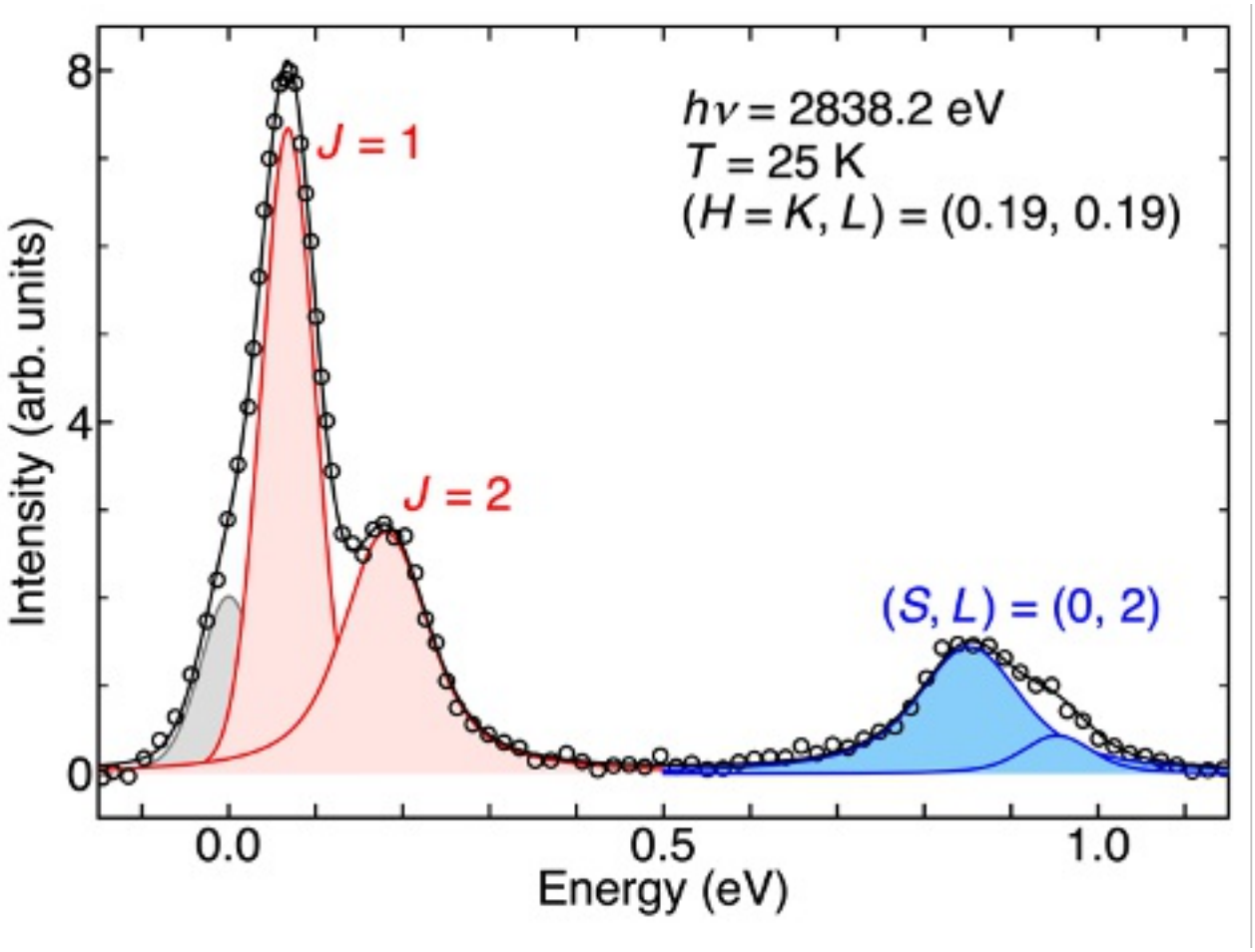

Fig. 1 Ru L_3 edge (2838.2 eV) resonant inelastic x-ray scattering spectrum of K_2RuCl_6 at a symmetric ($H = K$, L) = (0.19, 0.19) point of the Brillouin zone measured at 25 K. The spectrum is decomposed into five voigt functions corresponding to elastic scattering (gray), excitations to the spin-orbit J multiplets (red), and excitations to the (S, L) = (0, 2) states (blue).

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Charge density waves in infinite-layer $NdNiO_2$ nickelates

Charles Tam[1,2], Jaewon Choi[1], Xiang Ding[3], Stefano Agrestini[1], Abhishek Nag[1,4], Mei Wu[5], Bing Huang[6], Huiqian Luo[7], Peng Gao[5], Mirian García-Fernández[1], Liang Qiao[3], Ke-Jin Zhou[1]
[1]Diamond Light Source, Didcot, United Kingdom. [2]University of Bristol, Bristol, United Kingdom. [3]University of Electronic Science and Technology of China, Chengdu, China. [4]Paul Scherrer Institut, Villigen, Switzerland. [5]Peking University, Beijing, China. [6]Beijing Computational Science Research Center, Beijing, China. [7]Institute of Physics, Chinese Academy of Sciences, Beijing, China

Abstract

The record of high temperature superconductivity at ambient pressure is held by the cuprate family of superconductors. However, since their discovery over 30 years ago [1], the exact mechanism behind superconductivity in the cuprates still remains elusive. Since its first report in 2019 [2], infinite-layer nickelates thin films have emerged as a new manifestation of superconductivity with a cuprate-like structure. As well as the similar crystal structure, which is formed of NiO2 square planes, nickelates have the same undoped nominal $3d^9$ configuration with Ni^+ as the cuprates, and spin-waves are observed in the undoped parent compound [3]. There are notable differences, however, including participation of rare-earth orbitals at the Fermi level, and as of yet no evidence of long-range magnetic order nor other types of order, e.g., density waves as seen in cuprates [4].

In this work, we use resonant inelastic x-ray scattering at the Ni-L_3 edge to find evidence of a symmetry-breaking modulation, characteristic of charge density wave (CDW), in thin films of non-superconducting $NdNiO_2$. This CDW exists in Ni and Nd orbitals via

Ni-Nd hybridisation and has an in-plane modulation vector $\boldsymbol{Q} = (0.333, 0)$ reciprocal lattice units, with an in-plane correlation lengths $\xi = 20$ to 80 Å and non-negligible L dependence. By varying the growth conditions, we see a relationship between the CDW and the two-dimensionality of Ni and Ni-Nd hybridised orbitals. When superconductivity is induced with a substitution of Nd with Sr to form $Nd_{0.8}Sr_{0.2}NiO_2$, we see no evidence of CDW.

Our result highlights the ubiquity of CDW across the cuprate and nickelate superconductors, suggesting their importance to superconductivity.

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Resonant inelastic x-ray scattering study of the $CeAgSb_2$ Kondo system

B. Tegomo Chiogo[1], A. Chainani[2], D. Malterre[1]

[1]Université de Lorraine, CNRS, Institut Jean Lamour, F-54000 Nancy, France. [2]National Synchrotron Radiation Research Center, Hsinchu Science Park, Hsinchu 30076, Taiwan

Abstract

In cerium compounds, the strong electron-electron interactions of the f states and the hybridization of the f and conduction electrons lead to remarkable magnetic, electronic, and thermodynamic properties at low temperature such as heavy fermions, Kondo effect and intermediate valence [1,2]. Many high-energy spectroscopic studies such as core level and valence band photoemission spectroscopy (PES), inverse photoemission spectroscopy (IPES), X-ray absorption (XAS) have been devoted to determine the relevant energy scales (charge and spin excitations) in cerium compounds.

I will present our recent study on the ferromagnetic Kondo system $CeAgSb_2$ [3] by means of x- ray absorption spectroscopy and resonant inelastic x-ray scattering spectroscopy (RIXS) at Ce M_5 edge combined with simulation by using the single impurity Anderson model in the zero-bandwidth limit combined with full multiplet theory (QUANTY). We will show that all the charge excitations observed in combined PES and IPES can be evidenced by RIXS spectroscopy just by varying the incident photon energy around the Ce M_5 edge. We will also discuss about the drastic polarization dependence of the structure in RIXS spectra and the strong temperature dependence of the fluorescence-like structure which is reminiscent to the temperature dependence of the Kondo resonance.

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Author email: bodry.tegomo-chiogo@univ-lorraine.fr

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Probing the spin excitation spectrum of superconducting layered square planar nickelates

Sophia TenHuisen[1], Grace Pan[2], Denitsa Baykusheva[2], Jonathan Pelliciari[3], Stefano Agrestini[4], Dan Ferenc Segedin[2], Qi Song[5], Mirian Garcia-Fernandez[4], Abhishek Nag[6], Charles Brooks[2], Valentina Bisogni[3], Kejin Zhou[4], Mark Dean[7], Julia Mundy[2], Matteo Mitrano[2]
[1]John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, USA. [2]Department of Physics, Harvard University, Cambridge, USA. [3]National Synchrotron Light Source II, Brookhaven National Laboratory, Upton, USA. [4]Diamond Light Source, Didcot, United Kingdom. [5]Cornell University, Materials Science and Engineering, Ithaca, USA. [6]Paul Scherrer Institute, Villingen, Switzerland. [7]Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, USA

Abstract

A fundamental question in the physics of high-temperature superconductivity is the nature of the pairing interaction, with a spin-mediated pairing mechanism often proposed for the copper oxides. While the recently discovered nickelate superconductors share many structural and electronic motifs with the cuprates, it is unclear if the similarities extend to the role of spin fluctuations and the magnetic spectrum. To shed light on this question, we study the spin fluctuations of layered square planar nickelates Ndn+1NinO2n+2 with resonant inelastic x-ray scattering and band structure calculations. Doping of the nickel valence is achieved via structural tuning rather than chemical substitution, with superconductivity emerging in the n=5 sample for an effective nickel valence of d8.8. We investigate the magnetic excitation spectrum of the near-optimally doped, superconducting (n=5) square planar nickelate and

in the strongly overdoped, nonsuperconducting (n=3) compounds. Both compounds exhibit two weakly or nondispersing features, indicative of a substantial superexchange energy and consistent with measurements on doped infinite layer compounds as well as bulk La4Ni3O8 samples. Our experimental results indicate the presence of heavily damped spin fluctuations across the entire Brillouin zone and constrain the energy scales for the emergence of superconductivity in these materials in a spin-mediated pairing scenario.

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Ultrafast excitation of 4f orbital states - a new pathway to control magnetic order in metals

Nele Thielemann-Kühn[1], Tim Amrhein[1], Wibke Bronsch[1], Somnath Jana[2], Niko Pontius[2], Robin Y. Engel[3], Piter S. Miedema[3], Dominik Legut[4], Karel Carva[5], Unai Atxitia[1], Benjamin E. van Kuiken[6], Martin Teichmann[6], Robert E. Carley[6], Laurent Mercadier[6], Alexander Yaroslavtsev[6], Giuseppe Mercurio[6], Loïc Le Guyader[7], Naman Agarwal[6], Rafael Gort[6], Andreas Scherz[6], Siarhei Dziarzhytski[3], Günter Brenner[3], Federico Pressacco[3], Ru-Pan Wang[3], Jan Schunk[3], Mangalika Sinha[3], Martin Beye[3], Gheorghe S. Chiuzbaian[8], Peter M. Oppeneer[9], Martin Weinelt[1], Christian Schüßler-Langeheine[2]
[1]Freie Unsiversität, Berlin, Germany. [2]Helmholtz-Zentrum Berlin, Berlin, Germany. [3]Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany. [4]IT4Innovations-Czech National Supercomputing Centre, Ostrava, Czech Republic. [5]Charles University, Prague, Czech Republic. [6]European XFEL, Schenefeld, Germany. [7]European XFEL, Schenefeld, Ghana. [8]Sorbonne Université, Paris, France. [9]Uppsala University, Uppsala, Sweden

Abstract

Ultrafast magnetic response to optical excitation has been studied for many years; still, only very recently it has been appreciated that early electronic excitations may play a more important role than just being a transient step in the deposition of energy in the electron system [1,2]. In the case of 4f metals, f-f electronic excitations within the 4f shell are dipole forbidden. Therefore, a variation of the 4f electronic structure was generally assumed to be negligible for the ultrafast 4f magnetic response to optical excitations. From a recent RIXS experiment at FLASH 1 we learned that after exciting the 5d6s valence electrons in Tb metal with optical laser pulses, the 4f

electronic state actually can be affected on ultrashort time scales. The spin and orbital momenta derived from the 4f electronic state define the coupling of the magnetic system to the environment. According to our simulations of X-ray absorption (XA) data obtained from an XA experiment at the European XFEL, we find about 20% of the Tb atoms to be in an orbitally excited state [3]. The altered orbital state changes the magneto crystalline anisotropy. Our study gives a new dimension to the discussion about optically control of 4f magnetic dynamics, as it provides a femtosecond handle on the 4f spin coupling.

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Theory of Time-Resolved Resonant-Inelastic X-Ray Scattering in a Photoexcited Mott insulator

Takami Tohyama
Tokyo University of Science, Tokyo, Japan

Abstract

The recent development of time-resolved resonant inelastic x-ray scattering (TRRIXS) opens a new avenue for probing collective two-particle excitation, from which one can investigate novel photoinduced nonequilibrium phenomena in the wide range of momentum and energy spaces. RIXS can probe not only charge excitation but also magnetic excitation if one uses incident x rays tuned for L edge in transition metals. If the lifetime of an intermediate state in the L-edge RIXS process is short enough, the dominant contribution to the RIXS spectrum comes from the dynamical charge and spin structure factors. It is also numerically shown that even for a realistic lifetime scale of an intermediate state in cuprate materials, the magnetic excitation in RIXS gives information on the dynamical spin structure factor [1]. Therefore, TRRIXS is an ideal tool for characterizing transient spin dynamics.

We theoretically investigate momentum dependent spin excitation that evolves after pumping within a femtosecond timescale in the antiferromagnetic Mott insulator on a square lattice [2,3]. Using a numerically exact-diagonalization technique based on the time-dependent Lanczos method and time-dependent density-matrix renormalization group for a half-filled Hubbard model, we find novel momentum- dependent transient spin dynamics. In particular, we demonstrate characteristic temporal oscillations for the intensity of the dynamical spin structure factor, showing an antiphase behavior for two orthogonal directions that are parallel and perpendicular to the electric field of a pump pulse. The same behavior is also seen in the static spin structure factor. Their oscillation period in time is determined by two-magnon excitation in the Mott insulator. This theoretical prediction will be confirmed for Mott insulating cuprates

and iridates once TRRIXS is ready for a femtosecond timescale.

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The role of the ligand carboxylate electronic structure in a structural transition of a metal-organic framework (MOF) by oxygen K edge resonant inelastic x-ray scattering (RIXS)

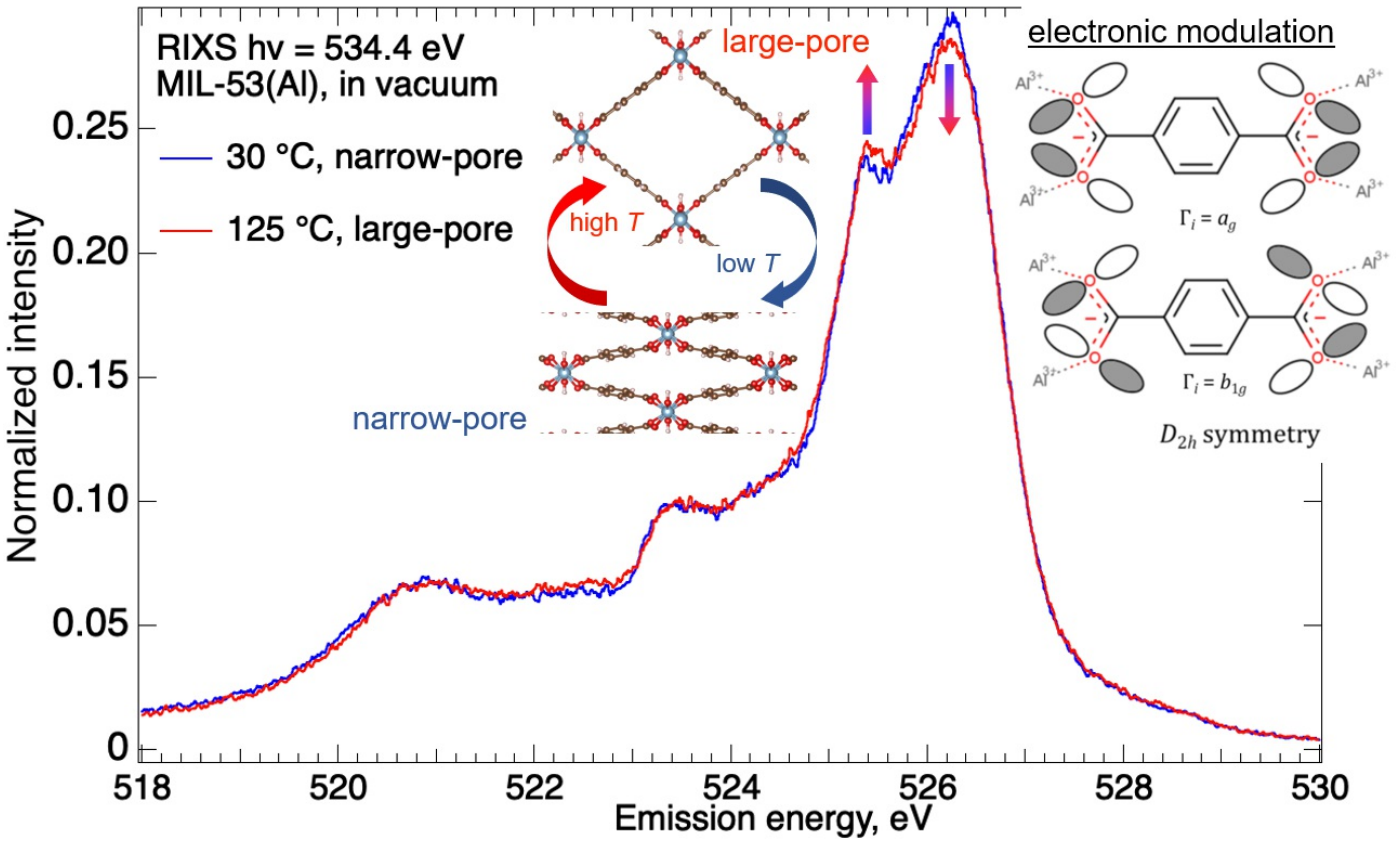
Ralph Ugalino[1,2], Kosuke Yamazoe[1], Jun Miyawaki[3], Naoya Kurahashi[1], Yoshihisa Harada[1,2]

[1]Institute for Solid State Physics, The University of Tokyo, Tokyo, Japan. [2]Department of Advanced Materials Science, The University of Tokyo, Tokyo, Japan. [3]Institute for Advanced Synchrotron Light Source, National Institutes for Quantum and Radiological Science and Technology, Sendai, Japan

Abstract

The remarkable structural flexibility of metal-organic frameworks (MOFs) in response to temperature and guest adsorption is aptly exemplified in the “breathing transition” of MIL-53 MOFs between narrow-pore and large-pore phases. Previous work suggested that the phase stability can be tuned by the interplay between van der Waals interactions and vibrational dynamics, with the narrow-pore phase, observed at low temperatures, and large-pore phase, observed at high-temperatures, being stabilized by enthalpy and entropy, respectively. We extend into the microscopic mechanism responsible for such thermodynamic behavior by focusing on the ligand carboxylate as the site of metal-ligand interaction which should be sensitive to changes in ligand dynamics responsible for the vibrational contribution to entropy. In this work, we used oxygen K edge resonant inelastic x-ray scattering (RIXS) to selectively probe the electronic structure at the linker carboxylate and observe the electronic modulation accompanying this structural transition. It was observed that the breathing transition solely induced by temperature involves a modulation of the occupancy of the carboxylate oxygen 2p lone pair orbitals occurring in either an antibonding- or a bonding-like configuration. The symmetry and near-degeneracy of these

electronic states opens the possibility of their coupling with vibrational soft modes that could drive certain distortions observed in the breathing transition. A two-level model was applied to estimate the contribution of such coupling to the reduction of force constant for such distortion and the thermodynamic parameters characteristic of such "mode softening". Finally, we note how water adsorption accompanying the breathing transition in ambient conditions involves additional perturbation of π states which suggests that a different mechanism could operate in the presence of adsorbed guest.



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Ferromagnetic resonance detected using soft x-ray absorption, reflection, and diffraction

Gerrit Van der Laan
Diamond Light Source, Didcot, United Kingdom

Abstract

Synchrotron radiation based techniques provide unique insight into both the element and time resolved magnetization behaviour in magnetic spin systems. X-ray detected ferromagnetic resonance (XFMR) has recently emerged as a powerful synchrotron-radiation-based tool able to study the element-selective magnetization dynamics [1]. Magnetic and chemical contrast in XFMR is obtained by x-ray magnetic circular dichroism (XMCD), while the phase difference between the magnetization precessions is monitored using stroboscopic probing. A unique property of time-resolved XFMR is the visualization of the magnetization precession for each individual layer in a magnetic device. Measurement of the amplitude and phase response of the magnetic layers gives a clear signature of spin-transfer torque (STT) coupling between ferromagnetic layers due to spin pumping.

We highlight the power of two recent developments, utilizing x-ray scattering techniques to reveal the precessional magnetization dynamics of ordered spin structures in the GHz regime, both in diffraction and reflection configurations. Our recently developed diffraction and reflectometry ferromagnetic resonance (DFMR and RFMR) techniques provide novel ways to explore the dynamics of modern magnetic materials, thereby opening up new pathways for the development of spintronic devices [2-5]. We provide an overview of these techniques and discuss the new understanding they provide into the magnetization dynamics in the chiral magnetic structure in Y-type hexaferrite and the depth dependence to the magnetization dynamics in a [CoFeB/MgO/Ta]4 multilayer.

Our characterization tools for the exploration of the dynamics of

chiral and multilayered magnetic materials are significant to the development of high-density and low-energy consumption data processing solutions.

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Resonant Inelastic X-ray Scattering at the uranium N-edge

Eleanor Bright[1], Lei Xu[2], Lottie Harding[3], Ross Springell[3], Martin Sundermann[4,5], Andrew Walters[6], Ping Yang[2], Enrique Batista[2], Abhishek Nag[7], Mirian Garcia-Fernandez[6], Ke-Jin Zhou[6], Gerrit van der Laan[6], Roberto Caciuffo[8], Gerry Lander[3]
[1]ESRF, Grenoble, France. [2]LANL, Los Alamos, USA. [3]University of Bristol, Bristol, United Kingdom. [4]Institute for Physics II, Cologne, Germany. [5]MPI for Chemical Physics of Solids, Dresden, Germany. [6]Diamond Light Source, Didcot, United Kingdom. [7]PSI, Villigen, Switzerland. [8]European Commission - Joint Research Centre, Karlsruhe, Germany

Abstract

Advances in the Resonant Inelastic X-ray Scattering (RIXS) technique using soft x-rays in recent years [1] have facilitated great advances in our understanding of 3*d* transition metal oxides such as the cuprates [2] as well as in rare-earth compounds (see for example [3]). It is currently uncertain whether soft x-ray RIXS could help provide similarly new insights into the physics of the actinides. To explore this possibility, we have performed RIXS studies on various uranium compounds (UO_2, α-U_3O_8 and UN) at the uranium N_4 and N_5 absorption edges using the I21 beamline [4] at Diamond Light Source. In UO_2, we have demonstrated that high-resolution RIXS can observe the crystal-field states (within the $^3H_4$ ground state) at around 150 meV, as well as the excited multiplet structure up to 1 eV [5]. The crystal-field states were measured with inelastic neutron scattering more than 30 years ago [6], but it was not possible to identify the higher states using neutrons. In the higher oxide α-U_3O_8, we have observed some of the multiplet features associated with a

$5f^1$ configuration with a ground-state of $^2F_{5/2}$ and the excited state of $^2F_{7/2}$, but we do not observe the expected strong transition at around 1.7 eV. We believe that this excitation is close to the band gap of α-U_3O_8 and is therefore broadened by additional possible transitions. In UN no sharp transitions have been observed, although the broad scattering response centred around 1 eV is considered a signature of a predominantly $5f^3$ configuration in this band-like semi-metallic system. We will discuss the implications of our results for these materials and discuss what the future may hold for RIXS studies of actinide materials.

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5

Identifying Cu-O and Cu-N coordination in NO_x removal: an *in situ* Valence to core-X ray emission spectroscopy study

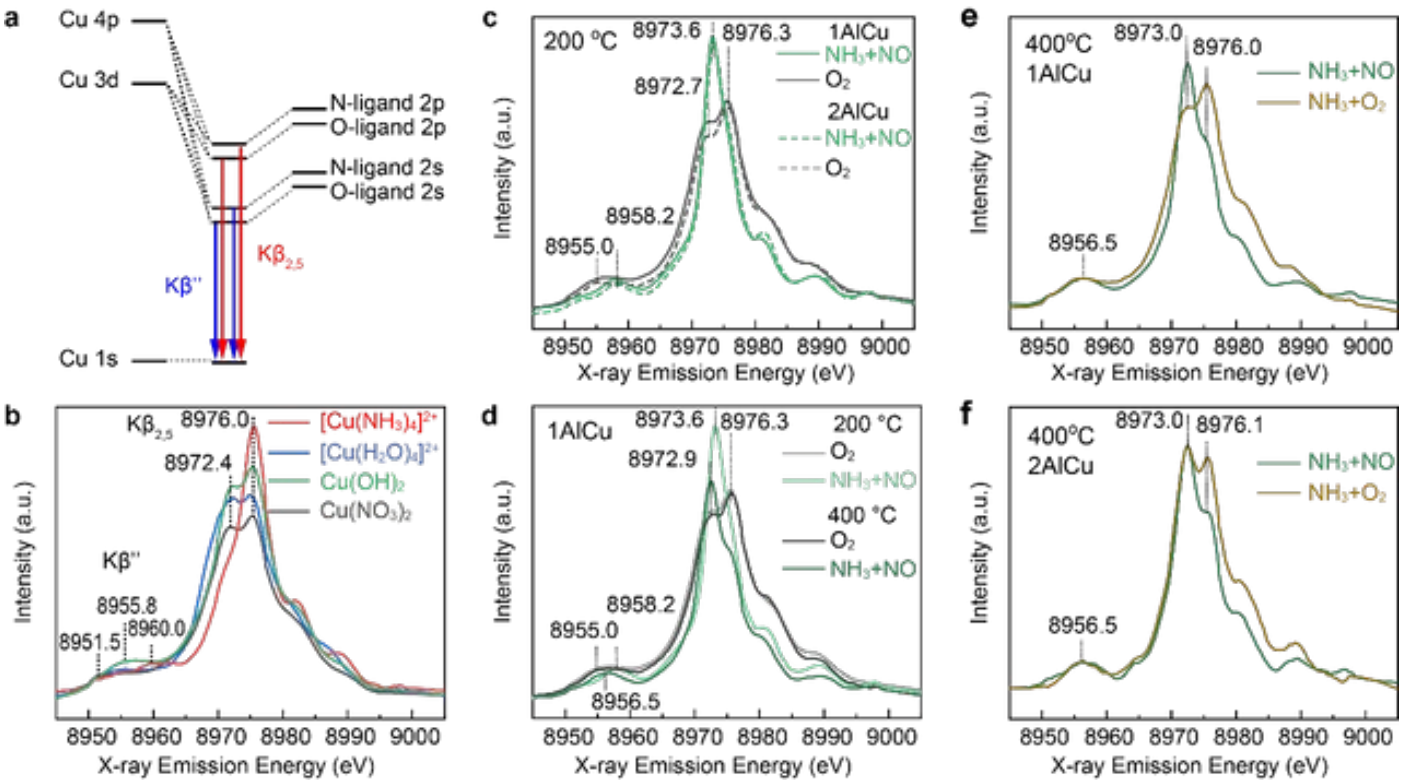
Yiyun Liu, Feng Ryan Wang
University College London, London, United Kingdom

Abstract

Cu-exchanged zeolite is an efficient catalyst to remove harmful nitrogen oxides from diesel exhaust gas through the selective catalytic reduction (SCR) reaction. The SCR activity is structure dependent, in which a Cu with one adjacent framework Al (1AlCu) has lower activation energy than Cu with two adjacent framework Al (2AlCu). Using a combination of operando X-ray absorption spectroscopy, valence to core - X-ray emission spectroscopy (VtC-XES)and density functional theory calculations, here we identified a $Cu^{I}(NH_3)_xO_{fw}$ (fw = framework) intermediate, the activation of which from Ofw-coordination is the rate-limiting step in the oxidation half-cycle and is significantly slower at 2AlCu site than 1AlCu site. As a result, the $CuI(NH_3)_xO_{fw}$ at 1AlCu breaks the Cu-O_{fw} coordination, enables the side-on coordination of O_2 and yields a transient $Cu_{II}NO_x$ intermediate upon interaction with NO. Our results demonstrate that controlling the strength of the Cu-O_{fw} coordination is key for high SCR performance.

In the SCR oxidative half cycle, the $CuI(NH_3)_2$ is oxidized to $Cu^{II}NO_x$ via $Cu^{I}(NH_3)_xO_{fw}$ intermediate. Such transition involves a change from Cu-N coordination to mixed Cu-O/N and finally to Cu-O coordination, which is studied with VtC-XES which is sensitive to the O and N–based ligands. The VtC spectra contain transitions to the Cu 1s core-level from the Cu 3d and 4p valence states that are mixed with ligand 2p ($K\beta_{2,5}$) and 2s orbitals (Kβ'' satellite) (Fig. 1a). O

coordinated $[Cu(H_2O)_6]^{2+}$, $Cu(OH)_2$, $Cu(NO_3)_2 \bullet 3H_2O$ have a double peak $K\beta_{2,5}$ feature at 8972.4/8976.0 eV while N coordinated $[Cu^{II}(NH_3)_4]^{2+}$ has a single peak at 8976.0 eV. The *operando* VtC-XES shows similar features for 1AlCu and 2AlCu samples at 200 ^{o}C (Fig. 1c), with $K\beta_{2,5}$ double peak for O-coordination at O_2 / NO + O_2 conditions, and N-coordination in the NH_3 related reductive conditions (Fig. 1c green,) The red shift of the $K\beta_{2,5}$ single peak from 8976.0 eV in $[Cu(NH_3)_4]^{2+}$ to 8973.6 eV in Cu-CHA suggests reduction to $Cu^{I}(NH_3)_2$. Increasing the temperature to 400 ^{o}C does not alter Cu coordination in oxidative conditions (Fig. 1d green), however, the double peak feature evolves in reducing conditions along with the red shift of Kβ'' to 8956.5 eV. This indicates the Cu-O coordination even under NH_3+NO for both 1AlCu and 2AlCu at 400 °C and is the key evidence for the $Cu^{I}(NH_3)_xO_{fw}$ feature. The $Cu^{I}(NH_3)_2$ to $Cu^{I}(NH_3)_xO_{fw}$ transformation is observed in changing from NO + NH_3 to NH_3 + O_2. In 1AlCu, the $K\beta_{2,5}$ feature under NH_3+O_2 is very similar to the initial fully oxidised state, indicating the oxidation to $ZCu^{II}OH$ (Fig. 1e). The 2AlCu maintains the Cu-N feature at 8973.0 eV, showing a mixture of Cu-N and Cu-O coordination (Fig. 1f). We conclude that 2AlCu contains a mixture of $Cu^{I}(NH_3)_xO_{fw}$ and Z_2Cu^{II}/$ZCu^{II}OH$ species. The complete conversion from $Cu^{I}(NH_3)_xO_{fw}$ to Z_2Cu and $ZCu^{II}OH$ at 2AlCu require the simultaneous presence of NO and O_2 for at least 1 hour.


a
Cu 4p
Cu 3d
N-ligand 2p
O-ligand 2p
N-ligand 2s
O-ligand 2s
Kβ''
Kβ2,5
Cu 1s
b
Kβ2,5
8976.0
8972.4
Kβ''
8955.8
8960.0
8951.5
[Cu(NH3)4]2+
[Cu(H2O)4]2+
Cu(OH)2
Cu(NO3)2
Intensity (a.u.)
X-ray Emission Energy (eV)
c
200 °C
8973.6
8976.3
8972.7
8958.2
8955.0
1AlCu
NH3+NO
O2
2AlCu
NH3+NO
O2
Intensity (a.u.)
X-ray Emission Energy (eV)
d
1AlCu
8973.6
8976.3
8972.9
8958.2
8955.0
8956.5
200 °C
O2
NH3+NO
400 °C
O2
NH3+NO
Intensity (a.u.)
X-ray Emission Energy (eV)
e
400°C
1AlCu
8973.0
8976.0
8956.5
NH3+NO
NH3+O2
Intensity (a.u.)
X-ray Emission Energy (eV)
f
400°C
2AlCu
8973.0
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8956.5
NH3+NO
NH3+O2
Intensity (a.u.)
X-ray Emission Energy (eV)

4

A setup for FLASH Liquid-phase UltrafastX-ray Spectroscopy (FLUXS)

Ru-Pan Wang[1], Martin Beye[2], Nils Huse[1]
[1]Hamburg University, Hamburg, Germany. [2]FS-FLASH, DESY, Hamburg, Germany

Abstract

The presented project aims at building a versatile liquid-phase spectroscopic instrument for advanced ultrafast solution-phase spectroscopy in the soft X-ray regime (< 1000 eV) to provide unique experimental opportunities to the worldwide user community at FLASH, the XUV and soft X-ray free-electron laser at DESY in Hamburg. By constructing a flexible system, in combination with beam splitting techniques for referenced detection, the end station is able to conduct (i) self-referenced X-ray absorption spectroscopy (XAS) with monochromatized X-rays pulses, (ii) pink-beam post-sample self-referenced dispersive X-ray absorption spectroscopy (dXAS) in transmission mode, (iii) partial fluorescence yield (PFY) absorption spectroscopy for low-concentration samples, and (iv) resonant inelastic X-ray scattering (RIXS) with moderate energy resolution across the whole photon energy range of FLASH.

The spectrometer aims for an intermediate energy resolution ($E/\Delta E \approx 1000$) for ultrafast molecular science, which allows for a compact and flexible design. It gives an opportunity to switch between transmission and scattering schemes. In transmission, measurements can be referenced by a using transmission grating beam splitter to create reference beams for Io measurements. Such a referencing scheme is vital for normalization at SASE XFELs with fluctuating spectral content and thus indispensable for dispersive transmission spectroscopy. We have successfully tested this experimental approach at FLASH using the two first-order beams of a soft X-ray transmission grating guided through or past a liquid flat jet, respectively [1].

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3

Dispersion damping of intermediate spin excitons in $LaCoO_3$ by thermal fluctuations

Ru-Pan Wang[1], Atsushi Hariki[2], Frank M. F de Groot[3], Jan Kuneš[4]
[1]Hamburg Univeristy, Hamburg, Germany. [2]Osaka Prefecture University, Osaka, Japan. [3]Utrecht University, Utrecht, Netherlands. [4]TU Wien, Vienna, Austria

Abstract

$LaCoO_3$, a compound exhibits multiple spin-state crossovers which coupling with complex magnetic and electric phase transition, where the charge, spin, and orbital degree of freedoms play an essential role. At low temperature, $LaCoO_3$ is a nonmagnetic insulator with Co ions in the low-spin (LS, S = 0) ground state. Upon heating, it undergoes a crossover to a paramagnetic Curie-Weiss insulator (T ~ 100 K). This spin-state crossover has been debated on the competition between the high-spin (HS, S = 2) and intermediate-spin (IS, S = 1) states for decades.

To address the question, we employ Co L_3-edge resonant inelastic x-ray scattering (RIXS) as an element specific with valence sensitivity technique to study the bulk $LaCoO_3$ across the thermally induced spin-state crossover around 100 K. Tomiyasu et. al. has shown that the Co L_3-edge RIXS is a unique method to study further details of the electronic structure on $LaCoO_3$ [1]. Owing to a high energy resolution of 25 meV, we observe unambiguously the dispersion of the IS excitations in the low-temperature regime [2,3]. Approaching the intermediate temperature regime, the IS excitations are damped and the bandwidth is reduced. The observed behavior can be well described by a model of mobile IS excitons with strong attractive interaction, which we solve by using dynamical mean-field theory for hard-core bosons [3]. Our findings refute the $LaCoO_3$ paradigm of the local atomic states interacting via spin exchange and replace it

with a picture of mobile strongly interacting excitons (IS) and bi-excitons (HS) on the LS background.

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Time-Resolved RIXS Theory and Potential Applications in Quantum Materials

Yao Wang
Clemson University, Clemson, USA

Abstract

The rapidly evolving quantum material research calls for precise control of collective electronic properties. Among various control knobs, the ultrafast laser pump is a promising approach due to the rich degrees of freedom and further motivates pump-probe spectral characterizations. Unlike equilibrium spectroscopies, ultrafast pump-probe spectra involve more theoretical complexity and are limited by more experimental constraints. In this talk, I will briefly introduce the theory of time-resolved resonant inelastic x-ray scattering (trRIXS). Then I will use correlated magnets as an example to show the applications in the ultrafast design of quantum matter from a theoretical and computational perspective. The rich information in trRIXS also allows recovering sub-resolution information, which is crucial for quantum information applications.

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RIXS for high energy batteries: demonstrations and challenges

Wanli Yang
Lawrence Berkeley National Laboratory, Berkeley, USA

Abstract

The demand of low-cost energy storage solutions with high capacity and high voltage performance has never been so keen due to today's energy applications, especially electric vehicles. Unfortunately, high-energy operations of battery devices often trigger detrimental effects that lead to performance decays. To some extent, understanding the decay mode of battery systems is even more critical than innovations of new materials and systems. This provides the opportunities for soft X-ray RIXS as a powerful characterization technique to reveal the relevant chemistry in battery materials.

This presentation focuses on the demonstrations of RIXS studies of transition metal-based battery electrodes. We argue that the high-voltage electrochemical operation drives oxide electrode systems into unconventional states that are often transparent with conventional X-ray absorption spectroscopy. In contrast, RIXS is capable of further resolve the entangled chemical states of both cations and anions, thus clarifying many misunderstandings of the performance issue of high energy electrode materials. In particular, we will discuss the novel metal states of anodes towards unusually low potential, and oxygen states of cathodes towards high potential. While recent researches have been trying to breakthrough some of the conventional wisdom on the understanding and employment of oxygen activities in battery cathodes, debates on both the fundamental mechanism and practicability of oxygen being a redox center remain active with controversy results and claims. Through soft X-ray RIXS, we try to clarify the different types of oxygen activities involved in cathodes charged to high voltages, which has been confused before and led to the misconception of oxygen redox

effects on performance [1]. We then discuss a comparative study of the oxygen activities in the so-called Li-rich, i.e., Li content higher than that in the typical stoichiometry, and conventional compounds to conclude that oxygen oxidation behaviors are not bounded to any specific Li configurations, instead, it is a common phenomenon among all transition metal oxide cathodes cycled to high voltages [2]. At the end, we argue that the oxygen states at high voltages in battery cathodes are unlikely from the unhybridized oxygen states, which dominates the models in battery field to understand high voltage performance. Instead, we found transition metals have a significant effect on the oxygen behaviors in battery cathodes, strongly indicating the key role of the metal-oxygen hybridization [3]. If time allows, we will present some of the material optimization strategy based on such fundamental findings from RIXS towards high energy battery electrode materials.

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Charge and spin excitations in quantum materials probed by RIXS

Ke-Jin Zhou

Diamond Light Source, Harwell Campus, Didcot, OX11 0DE, United Kingdom

Email: kejin.zhou@diamond.ac.uk

Resonant inelastic X-ray scattering is powerful for probing the strongly correlated quantum materials in which charge, spin, orbital, and lattice degrees of freedom are highly entangled. In this talk, I will firstly present our recent observations of charge density waves and acoustic plasmon excitations at the oxygen sites in hole-doped cuprate superconductors, $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ and $La_{1.84}Sr_{0.16}CuO_4$, revealing unusual behaviour of their valence electrons [1,2]. I will also flash through some of our works on the low-dimensional spin-chain systems, Y_2BaNiO_5 and $SrCuO_2$, where the higher-order magnetic excitations were only accessible by RIXS [3,4]. In the second half of my talk, our latest studies on the newly discovered infinite-layer nickelate superconductors $Nd_{1-x}Sr_xNiO_2$ will be presented in which magnetic excitations and charge density waves were found validating the existence of strong electron Coulomb interactions and symmetry breaking orders [5,6]. Finally, I will show RIXS studies on the flat-band Stoner excitations in a Kagome magnet [7].

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