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Speakers' Abstracts

High resolution RIXS from cuprates: results and perspectives

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We present a RIXS study of the low energy scale magnetic excitations in cuprates in the momentum resolved mode. These studies require high energy resolution in order to pick up the magnetic signal dispersing up to about 500 meV at the Brillouin zone boundary.

The starting point is the first evidence at Cu-L_3 edge of dispersion of bimagnons in antiferromagnetic cuprates obtained at the ESRF [1]. We present original RIXS results both on undoped and doped systems (mostly LCO and LSCO) obtained in the following conditions:
- at the Cu-L edge (measured at the SLS with the new ADDRESS beam-line equipped with the SAXES spectrometer [2])
- at the Oxygen K-edge (measured at the ESRF ID8 beam-line with the AXES spectrometer [3])
- at the Cu K-edge (measured at the ESRF ID16 beam-line)

A comprehensive discussion of the results at the various edges giving different sampling of the system allows a deep understanding of the magnetic excitations to be obtained. Moreover the comparison with neutron spectroscopy is illuminating.

The discussions shows also the present limitations mainly due to insufficient energy resolution in spite of the enormous progress done in the last couple of years. It is shown that a combined line-width not greater than 30 meV at 930 eV (Cu L_3-edge) is a realistic perspective in the coming years.

The results were obtained in various experiments involving many researchers who will be cited in the presentation. Without their contribution it would have been impossible to obtain the results presented here.

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Scattered photon polarization effects and symmetry analysis in "indirect" RIXS of cuprates

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As a symmetry-selective probe, Raman scattering has led to important contributions in the study of condensed matter systems. Limited to near-zero momentum transfer in the optical regime, this technique cannot yield information about the dispersion of electronic excitations. However, this is different for resonant inelastic X-ray scattering (RIXS), the analog of Raman scattering in the X-ray regime. Yet, little is known about the details of the RIXS cross section. More importantly, no consensus has been reached regarding the symmetry of the measured excitations or which selection rules, if any, these excitations obey. We present evidence of photon polarization effects at the Cu *K*-edge of several undoped cuprates that validates the use of polarization-based Raman selection rules to understand both the observed scattering geometry dependence and the photon polarization effects. We also show that, although excitations with A_{1g} symmetry dominate the RIXS spectrum, B_{1g} and E_g symmetric excitations are discernible.

From AXES to SAXES to …
Science and more in 15 years of Lucio's activity with x-ray emission spectroscopy

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Lucio Braicovich has been working for the ESRF and at the ESRF during the whole history of the facility, ever since the first proposal for such an ambitious facility. Lucio participated to the foundation of the ESRF, when he contributed to the famous "Red book" that contains the original scientific and technical case of the European synchrotron. He has been a member of the Scientific Advisory Committee for 8 years, and its chairman for 3. He has sit in several committees for the assignment of beam time. But at the same time, Lucio has never stopped coming to the ESRF as a very active user, submitting proposals, spending days and nights at the beam line, developing new instrumentation, analyzing data, writing and publishing articles for high level journals.

At 70 he is still getting fun with making experiments personally, designing new instrumentation, analyzing and organizing large numbers of spectra. And his enthusiasm for teaching is still as lively as before: he still spends hours and hours with new students for introducing them to the mysteries of experimental physics.

We will present an outlook of the scientific adventure of Lucio in the last 15 years, spent mainly working with resonant soft x-ray emission spectroscopy, first at ID12B, then at ID08 and recently also at the Swiss Light Source. Today the AXES spectrometer, designed in 1993, is still the second best in the world for energy resolving power in the 500-1000 eV energy range. Starting from that long and rich experience, SAXES was built for the ADRESS beam line of the Swiss Light Source. These two instruments have been opening totally new opportunities in the field of high resolution RIXS, resonant inelastic x-ray scattering. These new opportunities are to be explored in this workshop and are the scientific motivation of the new RIXS project for ID08, in the framework of the ESRF upgrade programme. Finally we will mention that Lucio has boosted two other important research fields at the ESRF, namely the pioneering photoemission spectroscopy made with hard x-rays and the so-called integrated resonant Raman scattering of x-rays.

20 years of inverse photoemission spectroscopy in Milano

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I shall review the major results obtained in our labs by Inverse Photoemission Spectroscopy, a technique introduced by Lucio Braicovich in Milano more than 20 years ago. The original system was based on a uv grating allowing for measurements at variable photon energies ($h\nu = 10 \div 25$ eV). The photon energy dependence of the (inverse) photoemission cross section has been extensively exploited for disentangling different orbital contributions to empty electron states in a wide class of solids, ranging from low *d* occupancy silicides and metallic alloys, to strongly correlated electron systems such as transition metals oxides and rare earth compounds.
Research was successively aimed towards magnetic materials: sensitivity to magnetic properties was achieved by realizing a spin resolved version of the technique, based on a spin polarized electron source and a band-pass photon detector working in the isochromat mode ($h\nu = 9.6$ eV). Empty electron states have been studied in a variety of magnetic systems, such as clean and oxidized surfaces, ultrathin films on metals, semiconductors and insulators, ferromagnetic-antiferromagnetic interfaces, half-metals, multilayers. A few cases, selected as representative of this wide research activity, will be discussed.
Finally I shall present most recent results on bi- and tri-layer systems based on Fe-NiO or MgO-Fe, which are of interest for applications in Exchange Bias devices and in Tunnelling Magnetic Junctions, respectively.

Invisible peaks and the relation between RIXS, fluorescence and Auger

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RIXS of localized systems is usually interpreted in a charge transfer multiplet picture. Transitions in other systems are interpreted with a band-like picture, for example studying elementary excitations in the final state. The level of interpretation is, as such, rather incomplete and in particular the localized systems have a range of additional features that are often 'invisible' (in XAS), not measured (yet) or omitted from the analysis. I discuss a few examples of invisible peaks and poorly understood phenomena.

(a) It is often assumed that electron yield and fluorescence yield detection result in identical x-ray absorption spectra, at least if experimental boundary conditions such as surface cleanliness and self-absorption effects are taken care of. A few examples are given where intrinsic differences arise between EY and FY, because of (1) energy dependent fluorescence yield cross sections (an effect that is very strong for nanoparticle systems [1]) and (2) broken coherence effects in fluorescence yield [2].

(b) The lanthanum L_3 edge in normal XAS has essentially a smooth edge structure. With 2p3d or 2d4p RIXS, the 2p5d quadrupole peak becomes visible [3]. Our recent data on 2p4d RIXS have shown additional peaks in the pre-edge region of the L_3 edge of La systems.

(c) The relative contributions of RIXS and normal fluorescence and variations of the normal fluorescence signal with energy vary dramatically from system to system. This is closely related to the situation in resonant photoemission (though apparently different in relative intensity) and relates to the issue of the changeover between the sudden approximation and the adiabatic limit.

(d) In 3d metal K edge RIXS, divalent oxides (in octahedral symmetry) have no (visible) dipole component in the pre-edge region [4], while oxides with higher valences have both visible dipole and quadrupole transitions, due to much stronger (oxygen mediated) metal-metal bonding [5,6]

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Table-top ultrafast coherent laser sources in the XUV: towards attosecond science

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The aim of the present work is to describe the ultrafast laser technology developments, which have proven to be crucial for the emergence of attosecond science. The generation of few-optical-cycle laser pulses of high energy in the near infrared with controlled electric field (carrier-envelope phase stabilization) can be considered as the first step. Two techniques for the generation of high-peak power few-optical-cycle pulses will be discussed, namely the hollow-fibre compression technique [1] and a particular implementation of the optical parametric amplification [2]. These pulses focused in noble gases can lead to the production of high order harmonics of the fundamental frequency radiation up to the soft X ray spectral region. In this spectral region attosecond pulses are generated.

Two schemes are presently used for generating attosecond pulses: (i) use of sub-6-fs driving pulses with controlled waveform and spectral selection of the cut-off portion of the harmonic spectrum [3]; (ii) the use of phase-stabilized few-cycle driving pulses in combination with the polarization gating technique [4]. Pulses comprised between 130 and 80 as have been generated using these methods.

Applications of attosecond pulses to ultrafast spectroscopy comprise: (i) the observation on the real-time of an atomic inner-shell process using a pump-probe experimental apparatus; (ii) the temporal dynamics of tunnel ionization: (iii) more recently the investigation of electronic processes in condensed-phase.

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Exploring the Dynamic Frontier of RIXS

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In materials science, the frontier of knowledge spans from molecular surface dynamics for heterogeneous catalysis with little explored transition states to functional materials with often surprising properties and phase transitions as well as to chemical dynamics in solution.
For these scientific questions we are now starting to apply RIXS as a probe of ultra fast dynamics with two complementary approaches. Using the femtosecond X-ray pulses from Free-Electron Lasers we probe the electronic structure dynamics of matter with femtosecond time resolved RIXS. Alternatively, we can study at sub-natural line width resolution wave packet dynamics on the intrinsic femtosecond scattering duration time scale of RIXS.

STEM-EELS characterization of individual nanostructures

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Scanning transmission electron microscope (STEM) and electron energy loss spectroscopy (EELS) have been widely used to characterize material at the nanoscale or individual nanostructures.
Recent instrumental developments such as the correction of the spherical aberration of electron lenses, the use of monochromator, the implementation of new signal treatment or of new CCD detectors have opened spectral domains. Spectroscopy with atomic resolution (<0.1 nm of spatial resolution) and energy resolution (<0.2 eV, energy domain from vis-uv to one keV) can now be accessed.
We will first give a brief introduction of the STEM-EELS techniques and discuss what are the main studied edges and the advantages and limitations of these techniques.
After, we will give several examples where nanostructures can be studied by such mean like :
- Metal nanoclusters where EELS can be used to investigate the optical properties [1],
- Transition metal oxide nanoparticles where EELS can monitor the local valency [2,3] at the 2p excitations, or d-d excitations at lower energy [4]
- Interfaces and multilayer where STEM-EELS can provide evidence for the nature of the terminating plane at the interfaces [5].

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Advances in X-ray detected magnetic resonance

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X-ray Detected Magnetic Resonance (XDMR) is a new spectroscopy in which X-ray Magnetic Circular Dichroism (XMCD) is used to *probe* the resonant precession of the magnetization when a strong microwave *pump* field $\mathbf{h}_p$ is applied perpendicu-larly to a static bias field $\mathbf{H}_0$. XDMR is element- and edge-selective and could become a unique tool to resolve the magnetization dynamics of spin and orbital components at various absorbing sites [1-3]. Typically, XDMR spectra recorded at K-edges reflect the precessional dynamics of pure *orbital* magnetization components.

We shall compare briefly different experimental configurations which we used to record Fe K-edge XDMR spectra in high quality yttrium iron garnet (YIG) thin films. Special attention will be paid in this presentation to non-uniform excitation processes under high pumping power [4]. We shall produce direct evidence that orbital magnetization components can couple to magnetostatic *spin waves* and that frequency doubling resulting from elliptic precession can be detected with XDMR. The latter experiments may open the door to the parametric amplification of orbital magnetization waves under the condition of parallel pumping.

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From the physics of metal-semiconductor interfaces to new physics with metal-semiconductor interfaces

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Understanding the chemistry and the electronic structure of metal-semiconductor interfaces in the initial stages of formation of the Schottky barrier has been one outstanding issue in surface science, of both fundamental relevance and practical interest. Through a pioneering use of photoelectron spectroscopy and synchrotron radiation, the Milano group – with multiple detours via Stanford – has made important contributions to this issue.

Two decades later metal-semiconductor interfaces are still the object of spectroscopic work as model systems where interesting physical effects of confinement and symmetry breaking can be tailored and studied, e.g. by high-resolution ARPES. I will discuss the lifting of the spin degeneracy predicted (Rashba effect) at surfaces, and the *giant* splitting recently observed by ARPES [1,2] in surface alloys, and in confined structures. These observations open new possibilities for the manipulation of spins without a magnetic field.

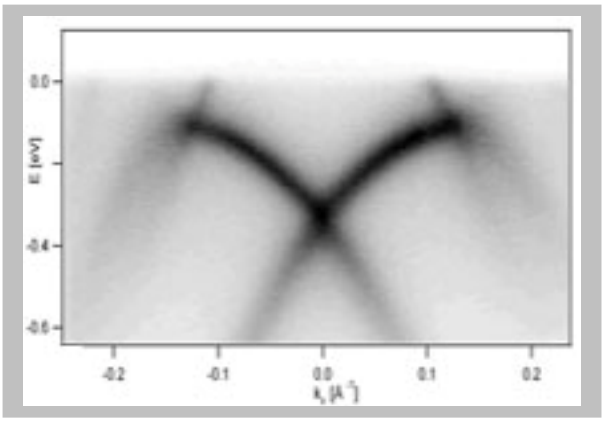

Figure 1: ARPES map of the spin-orbit split bands of a $BiAg_2$ surface alloy.

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RIXS to address novel condensed-matter physics

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Model calculations of resonant and non-resonant IXS studies on transition metal and rare earth compounds

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The theoretical description of most transition metal and rare earth compounds (strongly correlated materials) is still an enormous challenge. Traditional methods, like LDA using the independent electron approximation most of the time fail on even the simplest predictions. For example, many of the transition metal compounds, with NiO as the classical example, should be a metal according to band-structure calculations, but are in reality excellent insulators. One then has to resort to model calculations in order to gain understanding of these materials. The challenge being that most useful models one writes down cannot be solved, neither analytically nor numerically.

It thus is important to define experiments that can be theoretically understood in order to derive useful information about the ground-state properties of a material. X-ray absorption spectroscopy at the transition metal *2p* to *3d* or rare earth *3d* to *4f* edge has traditionally been a very successful technique. At these absorption edges the final states are strongly excitonic and therefore can be calculated with the use of multiplet ligand field model calculations. The understanding of the final state and selection rules then allows one to obtain information about the ground state. Even excitations within the optical gap, namely *d-d* or *f-f* excitations (orbitons / crystal field excitations) measured with either (resonant) inelastic x-ray scattering, neutrons or optics are also excitonic and have been described successfully with the use of multiplet ligand field calculations.

Recent developments in (resonant) inelastic x-ray scattering oppose a new challenge for theoretical model calculations. It now becomes feasible to measure *d-d* (*f-f*) excitations (orbitons) with such high resolution that one cannot approximate them as non-dispersive excitons. It is important to include the dispersion of the exciton as well as the interaction of the exciton with possible continuum states of different parity available at the same energy. For resonant x-ray scattering the resonant intermediate state is however still a strong exciton, which only can be solved due to its local nature. This then calls for model calculations where a different model and approximations for the intermediate resonant state is used then for the final state.

Within this talk I will introduce several model calculations that for the final state try to take into account the interaction between the local excitonic nature and the possible dispersion (magnon, orbiton) together with the interaction of these local states with band like states (with the appropriate band structure). For resonant spectroscopy I will introduce different models for the intermediate resonant state then for the final state. This allows one to make different approximations for the very different nature that these states might poses.

Hard X-ray RIXS studies of magnetic and orbital excitations

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Hard x-ray resonant inelastic x-ray scattering studies of copper-based compounds are reported. In the first part of the talk, I will focus on the use of Cu K-edge RIXS to probe the mid-IR region of the $La_{2-x}Sr_xCuO_4$ system. A new mode is observed around 500 meV. The doping, momentum, temperature and polarization dependence of this mode are reported. It is found to be strongly peaked at $(\pi,0)$, to be characteristic of the insulating parent compound, with a spectral weight that is rapidly suppressed on doping in carriers, and to be rapidly damped above room temperature, disappearing around 500 K. The excitation rotates the polarization of the incident x-ray and is only observed with that polarization perpendicular to the copper-oxide sheets. On the basis of these measurements, this mode is assigned to a two-magnon excitation. In the second half of the talk, I will discuss very recent measurements of the orbitally ordered system, $KCuF_3$. This work is motivated by the search for propagating orbital excitations known as "orbitons". By tuning to the Cu K-edge, orbital excitations were observed in the range 0.8 – 1.5 eV. The polarization and momentum dependence of these orbital excitations are reported.

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High-resolution RIXS with hard X-rays: advances and applications

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Resonant inelastic x-ray scattering (RIXS) in the hard x-ray regime has advanced greatly during the last few years. Both resolution and efficiency of RIXS spectrometers working in the several-keV range (including the *K* edges of 3d metals) have undergone improvements, allowing for instance an energy resolution of 100-200 meV for most resonances. The unique domain of hard x rays is true bulk sensitivity, allowing measurements also in complicated sample environments, e.g. in high-pressure conditions. The complementarity of hard x-ray RIXS with its sister technique, soft-x-ray RIXS, holds the promise of achieving a better understanding of the complicated electronic structure of strongly correlated materials when data from both spectroscopies are combined.

Resonant inelastic x-ray scattering in molecules: dynamical and polarization properties

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We combined experimental results and theoretical calculations on dynamical and polarization dependence properties of resonant inelastic x-ray scattering (RIXS) in gas phase after deep inner-shell excitation in the tender x-ray energy range (1-10 keV) of chlorine compounds [1, 2]. We will present here results obtained on Kα and Kβ emission from isolated HCl, CF_3Cl, CH_3Cl and Cl_2 molecules after resonant K-shell excitation (~2800 eV, core-hole lifetime of 1 fs).

Resonant Kα emission spectra have been recorded using our own x-ray emission spectrometer [3] at the Swiss Light Source, Villigen, Switzerland. RIXS spectra show that dynamical broadening of Kα lines caused by Franck-Condon distribution is quenched at the top of Cl (1s → LUMO orbital) dissociative resonance where the width of the Kα lines tends towards the core-hole lifetime. This behavior is explained by the parallelism between $1s^{-1}$ and $2p^{-1}$ potential energy surfaces calculated in our laboratory. It has been shown to be a general behavior. These effects are described in terms of resonant x-ray Raman scattering and dispersion and spectral shape of Kα lines are very well reproduced by our calculations. We will show that asymmetrical shape of the elastic structure is the signature of a nuclear motion in the intermediate excited state and depends strongly on the effective time duration of the scattering process. For small detuning, the elastic peak is broadened by the nuclear motion taking place on a sub-femtosecond time scale in the intermediate state. For large detuning, the wave packet has no time to spread out and a narrow structure is observed. This effect is reproduced by our theoretical approach: the wave packet theory of the resonant x-ray Raman scattering.

Moreover, we recently performed a study on the polarization properties of x-ray emission [3]. Measurements were done using the capability of the x-ray emission spectrometer setup of beamline 9.3.1 at the ALS [5] to analyze the polarization-dependence of RIXS spectra. A linear dichroism was observed which is a sensitive probe of molecular field effects.

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Magnetic circular dichroism effects in the resonant inelastic X-ray scattering at the K pre-edge of *3d* ions

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Transition metal elements play an essential role in physics, chemistry and geosciences. Properties such as magnetism, catalytic activity and optical absorption are governed by the *3d* orbitals, which can be probed by X-ray spectroscopies at the K pre-edge (*1s*→*3d*+*4p* transitions), such as X-ray Natural Linear Dichroism, Resonant Inelastic X-ray Scattering and X-ray Magnetic Circular Dichroism. Compared with the $L_{2,3}$ edges (*2p*→*3d* transitions), hard X-rays have several advantages: bulk sensitivity, the possibility to study samples in demanding environments such as high-pressure cells, and the smaller multiplet interaction between the core-hole and the *3d* orbitals, which enables to make a more direct connection to the ground state properties. However, XMCD effects are very small, due to the absence of spin-orbit coupling in *1s* core-hole. So far, this drawback hampered the interpretation of XMCD at the K-edge.

Here, we discuss a novel approach, which consists in coupling XMCD and RIXS spectroscopies at the K edge of *3d* ions. According to our preliminary calculations on Co (+II) within the multiplet method, large XMCD effects are expected in the *1s-2p* RIXS. This is very promising since the final state is the same as in $L_{2,3}$ absorption, indicating that RIXS-MCD could be an operative magnetic spectroscopy in the hard x-ray range. To confirm this prediction a more accurate description of the angular dependence in the multiplet calculation is required. This has been achieved by performing a spherical tensor analysis of RIXS.

As a first example of the method, I will present the case of *1s* → *3d* absorption. The spherical tensor analysis of XAS, followed by multiplet and DFT calculations has been used to study XNLD at the Cr K pre-edge in a $MgAl_2O_4:Cr^{3+}$ crystal. This approach has enabled us to fully interpret the dichroic signature of Cr^{3+} [1,2].

Then, I will provide a general angular dependence of the RIXS cross-section on the sample geometry and light-polarization. Within this new expression, the cross-section is written as a product of two terms, which can be determined by actual multiplet computational codes. Finally, we will derive the cross-section for the *1s-2p* RIXS with circularly polarized light (RIXS-XMCD).

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Two-electron photo-excited atomic processes near inner-shell threshold studied by RIXS spectroscopy

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In this work a high resolution resonant inelastic X-ray scattering (RIXS) spectroscopy was employed to separate spectral features pertaining to different two-electron atomic processes in the vicinity of an inner-shell threshold. The Johansson type crystal spectrometer coupled to the ID26 beamline of the European Synchrotron Radiation Facility (ESRF) was employed to record a comprehensive series of high energy resolution $KM_{2,3}$ and $KL_{2,3}$ spectra of Ar in the near-threshold region for 1s3p and 1s2p double excitations.

In case of 1s3p double excitations contributions of shake-off, shake-up, and resonant excitations were extracted from the Ar KM-$M_{2,3}$M X-ray satellite line intensity measured as a function of photon energy from [1s3p] double excitation threshold to saturation. The isolated [1s3p]nln'l' excitation spectrum is critically compared to the outcome of the multiconfiguration Dirac-Fock model with relaxation.

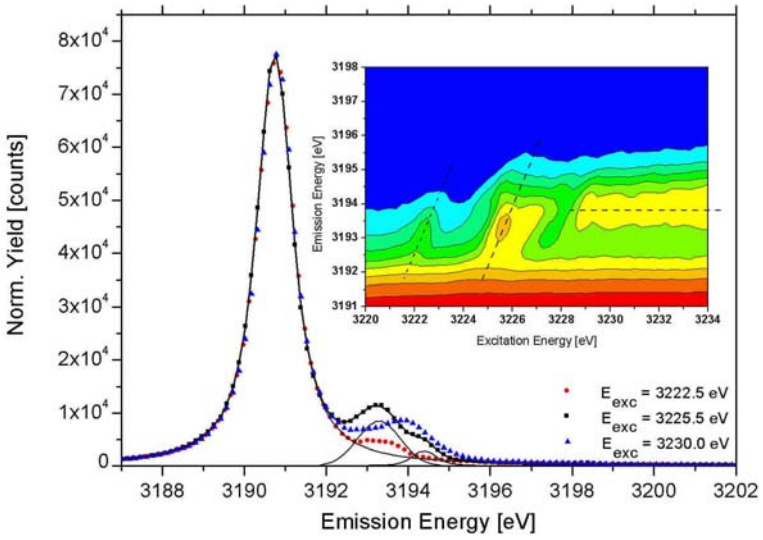

Figure 1: High resolution Ar $KM_{2,3}$ x-ray emission spectrum measured at three excitation energies close to the 1s3p threshold and contour plot represents a full RIXS scan over the 1s3p near-threshold region [1].

Despite that the 1s2p double excitation is dominated by the pure shake-off mechanism resonant contributions were clearly observed also at the 1s2p threshold. Since the separation from the dominating single-electron contribution is much more prominent than in absorption spectra the measured resonant emission spetrum provides an excellent base for comparison with the theory.

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Lucio Braicovich and RIXS – a look back and a look forward

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On the occasion of Lucio's 70 th birthday I will reflect on the first collaborative work we did exactly 30 years ago in 1979 at the Stanford Synchrotron Radiation Laboratory. The results were published the following year in three papers in Journal of Vacuum Science and Technology. The titles of the papers reveal what scientific issues we addressed: "Si-Pd and Si-Pt Interfaces", "Photoemission Investigation on the Temperature Effect on Si-Au Interfaces", and "Systematics of the Electron States of Silicon d-metal Interfaces". The work was all about using synchrotron radiation for photoemission studies of surfaces. This was the beginning of a long-standing collaboration between Lucio's group in Milano and mine at Stanford.
I will then try to convince Lucio what his research efforts should be focused on for the next 30 years. Notwithstanding the fact that Lucio's research on resonant inelastic x-ray scattering using synchrotron radiation has been both pioneering and has yielded a wealth of new information, I will point out that x-ray free electron lasers will open up even more possibilities. The Linac Coherent Light Source at Stanford achieved its first lasing in April 2009 at 0.15 nm just in time for Lucio's birthday – this cannot be an accidental coincidence! The laser will cover the photon spectral region 0.15 – 1.5 nm and can be tuned accurately around 2% of the binding energy for most of Lucio's beloved edges.

Infrared Synchrotron Radiation Spectroscopy at High-Pressure in Exotic materials

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The development of a metallic state from a correlated insulator, the interplay among competing energy scales in multifunctional systems, the appearance of delocalized quasi-particles across a Metal-to-Insulator transition represent paradigmatic examples of exotic behavior in modern Solid-State Physics. For instance in correlated systems, electronic delocalization is strongly reduced due to coulomb repulsion so that the interplay among competing spin, charge and lattice energy scales, might determine metallic states with different physical properties.
In general this interplay may be finely tuned using high-pressure and the corresponding changing in the low-energy electrodynamics can be tracked through infrared synchrotron radiation spectroscopy.
In this talk I will review the results that have been obtained using the infrared synchrotron radiation of the SISSI (Synchrotron Infrared Source for Spectroscopy and Imaging) beamline at Elettra on several families of exotic materials like Vanadium oxides, Charge Transfer Pyrite, and Graphite. Combining Infrared Spectroscopy results with the RIXS experimental technique may open interesting issues in the field of exotic and multifunctional materials.

Spin-excitations in cuprate Heisenberg models - RIXS with a neutron perspective

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We present recent results on spin excitations in Heisenberg ladders and square lattices, comparing the progress of neutron and RIXS studies. The neutron cross-section is long established, and experimental progress on e.g. the square lattice problem is limited by available flux and spectrometers. Understanding of the RIXS cross-section is in its infancy. We have found that excitations from the isotropic singlet ground state of spin ladders are two-triplets, whereas for the spontaneously symmetry broken antiferromagnetic state on the square lattice both single and two-spin-wave excitations are addressed, probably controlled by relative orientation of scattering vector, X-ray polarization and direction of the ordered moments. Further challenges include controling the Q-dependent intensity, where preliminary analysis indicate that even non-linear quantum effects can be seen with RIXS. While far from competing with state of the art neutron data resolution wise, RIXS can be performed on much smaller samples and may propel our understanding of cuprate quantum magnets, including the high-Tc problem.

RIXS Applied to Atoms and Molecules

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A short review on sub-keV resonant inelastic X-ray scattering (RIXS) applied to gas-phase samples will be given. Special attention on how core-hole localization, electronic-vibronic coupling, ultrafast symmetry breaking, and the dynamics during the ultra-fast scattering process are reflected in the spectra. Molecular RIXS is however heavily reliant on the availability of high brilliance photon sources, and so far the energy resolution has been compromised to attain reasonable count rates. New RIXS results associated with the double excitations in the helium atom measured at BESSY, and the 1s excitations of the oxygen molecule measured at the ADRESS bemline at SLS will be presented. The new generation synchrotron radiation sources facilitate measurements of spectra, in which individual vibrational peaks, much narrower than the lifetime width of the intermediate core-excited state, appear with good counting statistics. This implies a wealth of new opportunities, which will be briefly discussed.

IXS in correlated materials under high pressure

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Pressure is one of the most fundamental variables when dealing with structural stability and electronic properties of materials. But in spite of their fundamental interest, spectroscopic studies of materials under extreme conditions are difficult because of the strongly absorptive pressure sample environment.

As an all photon technique with considerable penetration power and a high sensitivity to the electronic states, IXS when performed in the hard x-ray range has proven to be most adapted to investigating the electronic properties under high pressure conditions. Both resonant and non-resonant IXS has provided in the last years considerable insights about the modification of the magnetic, valence, bonding or of the collective dynamics properties in materials with changing pressure (cf. review article, Ref. [1]).

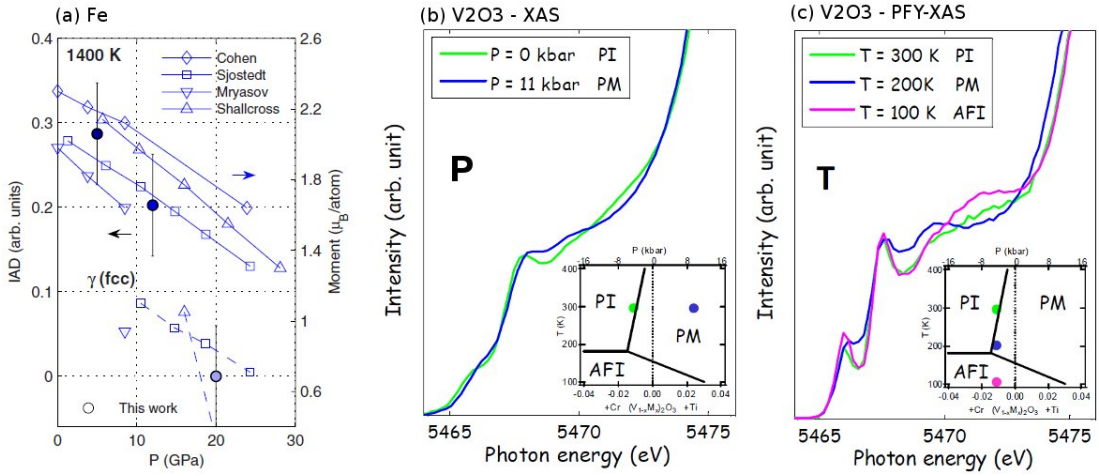

Figure 1: (a) Magnetic collapse in γ-Fe under pressure from (Ref. [2]); (b-c) V K-edge XAS at the pressure-driven Mott transition in V_2O_3 in transmission (b) and partial-fluorescence yield (c) modes.

We will present recent results of IXS under extreme conditions with an emphasis on *d* and *f*-electrons systems especially in strongly correlated materials. We will more particularly tackle the pressure-induced magnetic instabilities of iron at both low and high temperatures (cf. Fig. 1(a)), investigate the changes in the *d*-states through the Mott transition in V_2O_3 (cf. Fig. 1(b,c)), and discuss of the valence properties of rare earth and actinides under pressure and their consequences for electrons dynamics. Progress in the instrumentation and perspectives will be briefly reviewed.

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Resonant inelastic soft X-ray scattering in quasi one dimensional cuprates

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Resonant Inelastic X-ray Scattering (RIXS) is a powerful probe of excitations from the electronic ground state in transition-metal oxides. In the soft x-ray range the excitation energy can be tuned such that the photon scattering cross section with the partially occupied electron states is greatly enhanced. For example in studies with copper-oxides the charge, orbital and spin degrees of freedom of the Cu 3*d* states can be investigated by choosing the photon energy to the Cu $L_{2,3}$ edge (Cu $2p_{3/2,1/2} \rightarrow 3d$ transitions) [1]. In this talk we present high-resolution RIXS studies of magnetic and electronic excitations in the low dimensional spin systems $Sr_{14}Cu_{24}O_{41}$ and Sr_2CuO_3 performed at the ADRESS beamline of the Swiss Light Source with the SAXES spectrometer [2]. The experimental results are supported by calculations within an effective Hubbard model for a finite-size cluster.

$Sr_{14}Cu_{24}O_{41}$, which consists of two different copper-oxide layers: the so-called spin-ladders and spin-chains, is known to display a range of intriguing phenomena. Among others the chains develop a charge density wave at low temperatures, whereas the ladders possess a quantum mechanical spin liquid ground state with a finite spin gap [3]. With Cu L_3-RIXS we map out the dispersion of two-triplon excitations, the elementary magnetic excitations in the ladders, achieving excellent sensitivity over almost the full Brillouin-zone and directly determining the two-triplon energy gap [4]. Using O *K* RIXS, oxygen site-sensitive hole-excitations are studied, giving insight into the character of the holes in the ladders and the chains.

Sr_2CuO_3 is quasi one dimensional single-chain compound, which has the nearly ideal properties of a one-dimensional Heisenberg spin-1/2 system. It consists of corner sharing CuO_4 plaquettes forming chains along the crystallographic c-direction with a strong antiferromagnetic superexchange interaction around 0.26 eV between the neighboring Cu-sites mediated through a 180 degree Cu-O-Cu path. Following the dispersion of the Cu L_3-RIXS signal depending on the momentum transfer along the chain direction reveals that the main spectral weight follows the lower onset of the two-spinon continuum and that the magnetic RIXS spectrum is gapless at the Brillouin zone center. These results allow us to address the phenomenon of spin-charge separation in a one-dimensional spin-1/2 Heisenberg chain with RIXS.

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Determining crystal-field excitations in heavy fermion materials with x-rays

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Heavy fermion materials are strongly correlated rare earth or actinide materials where the atomic-like *f* electrons interact with conduction electrons giving rise to extraordinary low energy phenomena, such as unconventional superconductivity, competition between magnetism and superconductivity, and quantum critical behavior. Microscopic modelling of these phenomena starts with the understanding of the spatial distribution of the *f* electrons in the ground state and their local crystal-field excitations.

Neutron scattering has been up to now the standard technique to measure the crystal-field states in rare earth compounds. However there are limitations. Phonons and magnetic scattering, for example, typically occur in the same energy window, so that, when line widths are broad because of hybridization, it is difficult to separate the two. In addition absorption of some of the compounds' constituents (e.g. In, Rh or Ir in $CeRhIn_5$ or $CeIrIn_5$) causes problems in determining reliable magnetic intensities and in the case of the so-called giant crystal-field compound $CeRh_3B_2$ the energy splittings are just too large to be accessed with neutrons [1]. Recently we have successfully applied polarization dependent soft-x-ray absorption (XAS) to determine the crystal-field ground state wave function of several rare earth compounds [2], and now we are looking for new techniques to determine also the entire crystal-field excitations.

In resonant soft-x-ray scattering (RIXS) the electronic excitations are much stronger than phonons so that, as Ghiringhelli *et al.* have shown [3], it is a very promising tool to measure magnetic excitations. RIXS should therefore have the potential to also measure the crystal-field excitations in rare earths beyond the disturbing influence of phonons. Non-resonant inelastic x-ray scattering (NRIXS) has also a great potential for determining the *f-f* transitions by making use of higher multipole excitations. This was demonstrated by B.C. Larson *et al.* [4] for the case of the dipole-forbidden *d-d* excitations in NiO/CoO and explained quantitatively by Haverkort et al. [5] using a straightforward local many-body approach. The intensity can be optimized by varying the size of the transferred $|\mathbf{q}|$ while the vector $\mathbf{q}$ dependence of the excitations allows for a symmetry analysis as in polarization dependent experiments. An advantage of NRIXS over neutrons is that one can measure the local crystal field excitations over a very wide range of energies with good resolution, so that by looking at the higher multiplet excitations which lie above the phonon cut-off energy, one can obtain data free from phonons. Obviously, NRIXS is the best tool to determine the giant crystal-fields in $CeRh_3B_2$. In fact, the presently available resolutions should already be sufficient for a successful NRIXS and perhaps also RIXS study on this particular compound. Upon improvement of the energy resolution and the detector collection angle in RIXS and NRIXS, one could dream about entering a research field which at the moment is dominated by neutrons thereby also obtaining new information which is not accessible so far.

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Inelastic soft X-ray scattering of protein and amino acids in water

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Soft X-ray RIXS is a powerful tool for studies of electronic structure of various materials, such as semiconductors, ionic materials, simple metals, and strongly correlated materials[1]. Furthermore, Soft X-ray RIXS is also powerful for the liquids and wet samples. Electronic structures of these materials are very challenging and can be expected to provide important results in the near future. We will discuss about the electronic states of transition metal ions in proteins in water, which play important roles in protein-function. It is well known that met-Myoglobin has a high-spin (Fe^{3+}) and CO Myoglobin has a low-spin (Fe^{2+}) configuration. By changing the spin and electron valence, Myoglobin can bind or release various gaseous species such as oxygen, nitrogen, etc. We have measured the Fe 2p resonant- soft X-ray RIXS spectra of Myoglobin, which show characteristic d-d transitions corresponding to spin and valence configuration of the transition metal ions.[2]

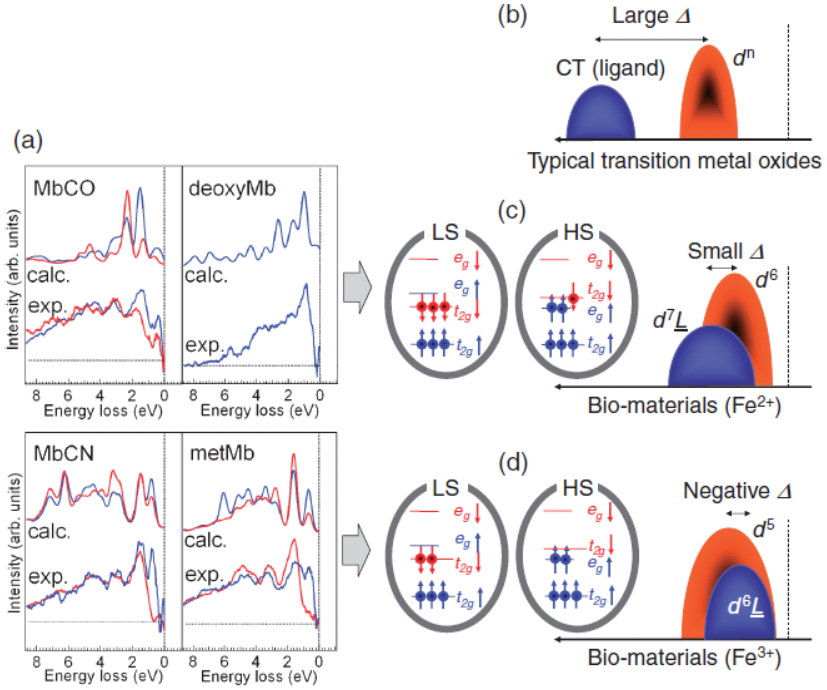

Figure 1 (a) Experimental and theoretical results for Fe $2p_{3/2}$ RXES of MbCO, deoxyMb, MbCN, and metMb. (b)–(d) Schematic energy diagram for $3d^n$ system corresponding to different cases; (b) A model typical for transition metal oxides, characterized by a large positive charge transfer energy Δ, the energy separating the atomic-like d^n-states and the ligand (CT) states; (c) A model for (Fe^{2+}) myoglobin characterized by a small positive charge transfer energy Δ; (d) A model for (Fe^{3+}) myoglobin characterized by a negative charge transfer energy Δ.

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The light that resonant Inelastic X-ray scattering throws on High Tc cuprates

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Resonant Inelastic X-ray Scattering (RIXS) provides direct access to elementary charge, spin and orbital excitations in complex oxides. As a technique it has made tremendous progress with the advent high-brilliance synchrotron X-ray sources. The figure of merit for K-edge RIXS experiments on the cuprates (inelastic count-rate/energy resolution) has for instance improved by four orders of magnitude in the last ten years.

Theorists in the mean time, asked themselves to which low-energy correlation functions RIXS is precisely sensitive. I will show that depending on the experimental RIXS set up, the measured charge dynamics can include charge-transfer, d-d and orbital excitations [1,2,3]. RIXS also allows to probe spin dynamics, in particular the dispersion of magnons and bi-magnons [4,5]. Based on these observations, I will discuss the novelties that RIXS can reveal on the spin dynamics of High Tc cuprates, particularly at the copper L-edge [6].

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Analysis of magnetic circular dichroism in resonant Raman scattering using transverse geometry

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Results are presented for resonant Raman scattering (RRS) in transverse geometry excited with circularly polarized x rays at the Co and Ni $L_{3,2}$ edges in $CoFe_2O_4$ and $NiFe_2O_4$ and in Co and Ni metal. [1] In this geometry the incident x-ray beam is perpendicular to the magnetization and the magnetic circular dichroism (MCD) is only present in the RRS and not in the absorption. The work is based on the measurement of the spectra vs the incident photon energy and integrated over the scattered photon energy. In all samples the 3*s*-RRS channel terminating in an inner shell 3*s* hole and extra 3*d* electron ($2p^6 3d^n \rightarrow 2p^5 3d^{n+1} \rightarrow 2p^6 3s^1 3d^{n+1}$) is about an order of magnitude larger than the 3*d*-RRS channel ending in a 3*d* electron-hole pair ($2p^6 3d^n \rightarrow 2p^5 3d^{n+1} \rightarrow 2p^6 3d^n$). The result can be explained by a theory based on the fast-collision approximation.

Integrated resonant Raman scattering (IRRS) is ideally suitable for sum-rule analysis [2]. In magnetic systems it gives ground state information as represented by an expansion in coupled multipoles of orbital and spin moments up to order 4. Therefore, applying the sum rules characteristic of a second-order process IRRS gives access to moments higher than those obtained from magnetic circular and linear dichroism in x-ray absorption [3]. The experimentally deduced values for the quadrupole and octupole moments are small in Ni ferrite but large in Co ferrite [4]. Except for the spin moment, this difference between both ions is in contrast with the ground-state values given by the Hund's rule that gives the same absolute value for both isolated ions. The difference is due to the quenching of the spin-orbit interaction (its first-order contribution vanishes) in the octahedral crystal field for the Ni d^8 ion, which does not occur for the Co d^7 ion. The results show the extent to which the atomiclike properties are retained in the higher-order multipole moments. This can be used to assess the modification of the atomic behavior due to solid-state effects.

Measurements of the MCD in IRSS in the perpendicular geometry for the $L_{2,3}$ absorption region in Fe, Co, and Ni metal show that spin-dependent screening of the core hole takes place within the scattering time scale, which is supported by the absence of the effect in ionic systems [5]. This allows an assessment of the time scale for the screening process (up to a few femtoseconds). The process is almost complete within the scattering time for Fe and Co metal, but this is not the case for the narrow band metal Ni which shows a much slower dynamics. Quantitative results can be obtained using a linear rate model [6].

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Fine details of cobalt 1s pre-edges revealed by RXES

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The metal 1s pre-edges in compounds of 3d transition metals are rich in information, as they can reflect valence and spin state, coordination number and local symmetry simultaneously. However, often there are several obstacles to address the fine details of the pre-edge, including its low intensity, the poor separation from the tail of the main edge, and the large overlap and smearing due to the large lifetime broadening of the 1s core hole in the XAS final state. Resonant X-ray emission spectroscopy (RXES) can overcome these difficulties and can unveil the details of the underlying transitions. In this presentation we wish to show intriguing examples of RXES applied to cobalt oxides, which demonstrate that resonant techniques are often indispensable to pre-edge studies. These include the case of Co_3O_4, where the resonances of different sites happen to appear at the same incident energy; $LaCoO_3$, whose deceptive spectral shapes in the pre-edge often lead to a faulty assignment; and non-local (or off-site) transitions, that gauge intersite p-d hybridization between neighbouring metal atoms.

In addition, the exceptional resolving power of the presented 1s2p RXES experiments allows us to demonstrate the coherent second-order nature of the underlying scattering process. A relevant part of the talk is covered in Ref. 1.

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Intra-site electronic correlations in polarization-resolved resonant inelastic soft X-ray scattering

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Understanding the nature of electronic excitations in correlated materials and their signature in Soft X-ray spectroscopies requires a systematic way of modeling them. We present here some results for polarization-resolved RIXS where spectra are calculated within a multiplet crystal-field model. With specific examples calculated with our code, we show that intra-site correlations and excitations can be well characterized with the help of polarization-resolved experiments. The effect of inter-site correlations and their emergence in the spectra will also be discussed.

RIXS study on high-Tc cuprates and related compounds

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We will report recent results of RIXS study on high-Tc cuprates and related strongly-correlated electron compounds carried out searching for charge excitation associated with the charge order in these compounds.

In the strongly-correlated electron systems, charge ordered states are often realized as a results of the strong correlation. Particularly, in the high-Tc cuprates, charge stripe states are realized in the superconducting region and dynamical version of the charge stripes are expected to be related to the superconductivity.

We have carried out RIXS study in the hard x-ray regime for materials $La_{2-x}Ba_xCuO_4$ (LBCO) (x=0.125) and $La_{2-x}Sr_xNiO_4$ (x=1/3). Both materials are well known to have charge ordered state in the form of stripes, but they have different geometry: the former shows parallel stripes and the latter shows diagonal stripes. In both materials, we observed additional RIXS spectra near 1 eV at the momentum transfer Qs corresponding to the stripe spatial period. In this talk we will introduce briefly RIXS studies of other charge ordered cuprates and will discuss the excitations that is universal to the charge ordered state of the strongly correlated electron systems.

This study is carried out in collaboration with K. Ishii, K. Ikeuchi, J. Mizuki, K. Kakurai (JAEA), H. Kimura, M. Fujita, Y. Noda, K. Yamada, T. Adachi, Y. Koike (Tohoku Univ.), A. H. Said, and Yu. Shvyd'ko (ANL-APS).

The metal-insulator-transition in VO_2 investigated by resonant inelastic X-ray scattering

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Vanadium dioxide VO_2 undergoes a metal-to-insulator transition (MIT) from a paramagnetic metal with tetragonal rutile structure to a nonmagnetic semiconductor with monoclinic structure below 340K, characterized by a sudden conductivity drop of several orders of magnitude [1]. The crucial question in the ongoing debate on the nature of this MIT is, if the band gap opening is dominated by the crystallographic distortion (Peierls-transition) [2] or electron correlation effects (Mott-transition) [3]. We investigate changes of the electronic structure across the MIT in VO_2 by temperature dependent Resonant Inelastic X-Ray Scattering (RIXS) at the ADRESS beamline of the Swiss Light Source. RIXS is a unique photon-in / photon-out spectroscopic probe for determining the energy and symmetry of charge neutral excitations (e.g. crystal field or spin excitations) in strongly correlated materials. Our high-resolution RIXS study at the V L_3-edge reveals clear changes in the crystal field originating from the crystallographic distortion accompanying the MIT, especially a remarkable electronic excitation at -0.45 eV broadens and nearly merges with the elastic peak when going from the insulating to the metallic phase. Furthermore, we investigated the temperature dependent hysteresis of this excitation [4]. Differences between experiment and simulation within multiplet theory are discussed in relation to screening and charge fluctuation effects.

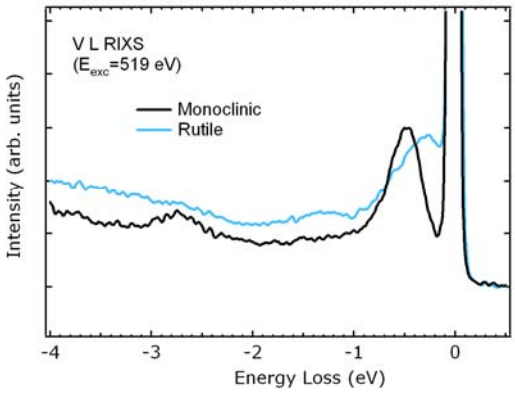

Figure 1: RIXS spectra of VO_2 both phases excited at the incident energy 519 eV.

Refereces

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