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Magnetic Resonant Inelastic X-ray Scattering

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Aristotle attributes in *De Anima* the first discussion on magnetism to Thales of Miletus, who lived in Asia Minor, present-day Turkey, at around 600 BC. It is remarkable that despite its ancient roots, research on magnetism and magnetic materials has remained a pillar of condensed matter physics and materials science, and continues to go from strength to strength. Other scientific ideas pioneered by Thales of Miletus, such as his cosmological thesis which held that the world started from water, have gained considerably less momentum.

The aims of present-day applied research on magnetic materials range from improving solid-state data-storage and read-out devices to the development of spintronics, which envisages an entirely new form of computation based on the manipulation of spin currents instead of charge currents. The scope of basic research on magnetism is possibly even broader. In for instance frustrated magnets, characterized by strongly competing interactions between spins which prevents the onset of magnetic ordering even at the lowest temperatures, novel types of spin-liquid states are being explored. Another hot topic is unraveling the relation between magnetism to superconductivity, in particular the role of magnetic interactions in mediating the pairing of electrons, a field boosted by the discovery of high-temperature cuprate superconductors in the late 1980's and of iron-pnictide based superconductors just a few years ago.

In order to establish how the magnetic properties of solids can be used and manipulated, it is essential to understand the spin dynamics of a magnetic material. Whereas static magnetic properties are determined by the magnetically ordered (or disordered) ground-state, an understanding of spin dynamics requires fundamental knowledge on the magnetic excitations of the system. The magnetic ground-state can be viewed as the "magnetic vacuum" of the system and any magnetic excitation as a "particle" created on top of this vacuum. Such quasi-particles are, e.g., the magnons in ferro- and antiferromagnets, or the spinons in quasi one-dimensional spin-chain compounds. The dispersion of these quasiparticles, which relates their energy to their momentum, and the interaction between them, entirely governs the spin dynamics.

Measuring these magnetic quasi-particles, in particular their dispersions, has traditionally been the exclusive domain of inelastic neutron scattering (INS). In the past few years, however, Resonant Inelastic X-ray Scattering (RIXS) has started to challenge this monopoly. INS can probe with great precision the lower energy magnon dispersions and, more generally, the dynamic magnetic structure factor $S(\mathbf{q}, \omega)$ of ordered and disordered magnetic systems, where $\mathbf{q}$ is the momentum and ω energy. It is a very powerful and mature technique, but also has systematic difficulties. As a rule INS requires large sample volumes so that measuring $S(\mathbf{q}, \omega)$ on for instance very thin films or other nanostructures is extremely challenging. Another, less generic, drawback is that the possibility to do INS on materials containing neutron absorbing elements (e.g. Cd, Gd, Ir or Os) is rather limited.

RIXS is a fast-developing experimental technique in which one scatters X-ray photons inelastically off matter, see Fig. 1. It is therefore a *photon-in − photon-out* spectroscopy for which one can, in principle, measure the energy, momentum, and polarization change of the scattered photon. The change in energy, momentum, and polarization of the photon are transferred to intrinsic excitations of the material under study and thus RIXS provides information about those excitations. RIXS is at the same time a resonant technique in which the energy of

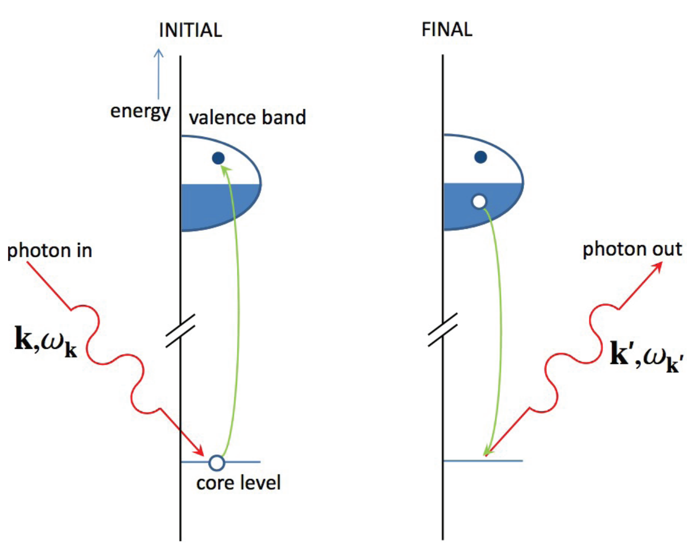


Figure 1: In a RIXS process the incoming X-rays excite an electron from a deep-lying core level into the empty valence [2]. The empty core state is then filled by an electron from the occupied states under the emission of an X-ray. This RIXS process creates a valence excitation with momentum $\hbar q = \hbar \mathbf{k}' - \hbar \mathbf{k}$ *and energy* $\hbar\omega = \hbar\omega_{\mathbf{k}} - \hbar\omega_{\mathbf{k}'}$.

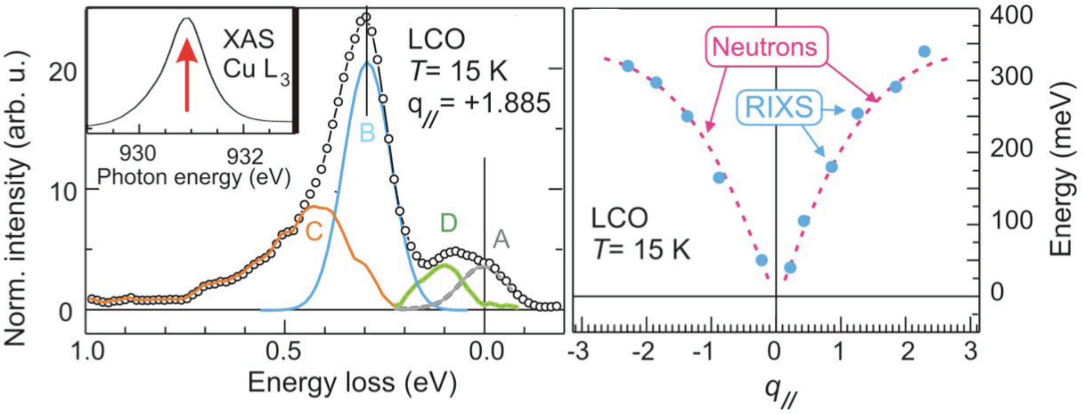


Figure 2: Left: RIXS spectrum at the copper L_3 edge at 931 eV of La_2CuO_4 with an energy resolution of 140 meV [7]. The transferred momentum (projected on the copper-oxide plane) is along the (π, 0) direction. Feature B, the maximum of the spectrum, is the magnon signal. Right: Comparing the resulting magnon dispersion measured by inelastic neutron scattering and RIXS at the copper L_3 edge of La_2CuO_4 .

the incident photon is chosen such that it coincides with, and hence resonates with, one of the atomic X-ray transitions of the system, depending on the edge in either the soft or hard X-ray regime. The resonance can greatly enhance the inelastic scattering cross section, sometimes by many orders of magnitude, and offers a unique way to probe, in principle, charge, magnetic, and orbital degrees of freedom on selective atomic sites in a crystal [1, 2].

Inelastic *neutron* scattering relies on the spin $\hbar/2$ of an incoming neutron interacting with an electron spin in the solid, via the magnetic dipole interaction, which allows for a simultaneous spin-flip of neutron and electron. The scattered neutron then carries the information on the energy and momentum of magnetic excitations left behind in the solid. The magnetic exchange interaction between a neutron and an electron spins that generates the spin-flip is local, but the excitation created in the scattering process, for instance a magnon, can of course be delocalized and wave-like in nature, being characterized only by the momentum $\mathbf{q}$ and energy ω transferred from the neutron to the magnetic quasi-particles in the spin-system.

Obviously the same kind of magnetic scattering can be envisaged with X-ray photons, as a photon carries an angular momentum of $\hbar$ that can in principle be transferred to the electron spin-system, causing magnetic excitations in the solid. However, the direct interaction between the photon and the spin via the magnetic part of the electromagnetic field – a Zeeman type of interaction – is extremely small, which prohibits straight-forward experimental implementation of this idea.

The way around this extremely small *direct* X-ray-electron spin scattering cross-section is to tap into an alternative route of transferring the photon angular momentum to the electron spins, via the very strong spin-orbit coupling of core electrons. Precisely this scattering channel is exploited by magnetic RIXS, where by the initial absorption of the X-ray photon, an electron is promoted out of the atomic core to the valence band. In the ensuing intermediate state the core-hole with a spin of $\hbar/2$ experiences a very strong spin-orbit coupling and if the core-hole orbital is of p character and thus has an angular momentum of $\hbar$, the core-hole can exchange part of its angular with its spin momentum, thereby flipping the spin of the core-hole.

Due to the very large spin-orbit coupling in the atomic core – about 20 eV for a Cu $2p$ core-hole, for instance – the probability for spin-flip of the core-hole during the intermediate state is substantial: it is of the same order of magnitude as the probability for a non-spin-flip of the core-hole. (It is actually rather straight-forward to calculate the cross sections for these two processes. Which one is larger depends on the precise scattering geometry and X-ray polarizations that are used.) After a core-hole flipping its spin, the electron originally promoted to the valence band cannot decay and fill the core-hole, as it now has the wrong spin. However, any valence electron with the opposite spin can decay and fill the core-hole and in doing so emit the out-going X-ray. Even if this perhaps appears to be a complicated process, with a lot of action in the intermediate state, the net-result is very simple: in magnetic RIXS a valence electron flips its spin, precisely as it would have when interacting with a neutron via a magnetic-dipole interaction, creating an elementary magnetic excitation, a magnetic quasi-particle, in the spin-system.

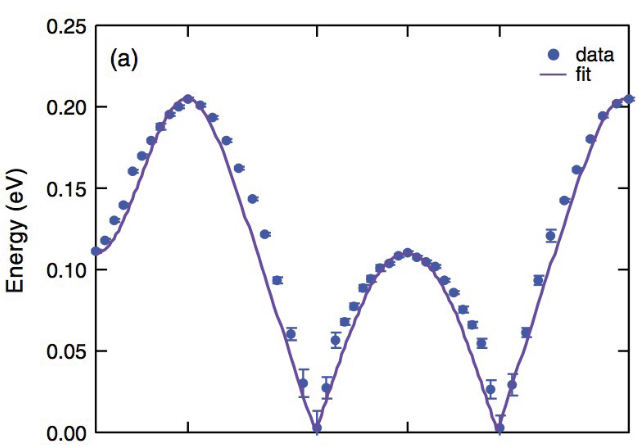
So in RIXS it is in principle possible for a X-ray photon to couple to the electron spin-system via the strong spin-orbit interaction in the atomic core. It turns out, however, that there are very strong selection rules. If one considers for instance RIXS at the copper L-edge, which starts by an initial Cu $2p \Rightarrow 3d$ transition and is followed by a $3d \Rightarrow 2p$ decay, it is not always possible to make a pure spin-flip excitation. In particular for a system containing Cu^{2+} in a $3d^9$ configuration with the Cu hole in a $x^2 - y^2$ orbital – the most common situation in copper-oxides – it is not possible to reverse the spin in a $x^2 - y^2$ orbital if the spin is initially pointing along the z-axis: $x^2 - y^2_\uparrow \not\Rightarrow x^2 - y^2_\downarrow$. In a seminal paper De Groot and coworkers [3] have shown that in this situation the only possibility to reverse the electron spin is by changing at the same time the orbital state of the electron, for instance from $x^2 - y^2$ to $3z^2 - r^2$ so that $x^2 - y^2_\uparrow \Rightarrow 3z^2 - r^2_\downarrow$ is allowed in RIXS, but pure spin-flip (without accompanying orbital excitation) is not [4]. Because of this observation, it was believed for a long time that the potential of pure magnetic RIXS was, at least for cuprates, not very big, even if it would be possible experimentally to reach the very challenging goal of a ω-resolution on the order of 0.1 eV or better, which is needed to discern the magnon dispersion in a typical copper-oxide antiferromagnet such as $La_2\,CuO_4$.

All this changed in 2009, when it was pointed out that the situation becomes very different when the Cu spin does not point along the z-axis, but is oriented in the xy-plane [5], which coincidentally is the case in most magnetic cuprates. For such a spin orientation the RIXS matrix elements turn out to be such that $x^2 - y^2_{\leftarrow} \Rightarrow x^2 - y^2_{\rightarrow}$ is an allowed, and very strong, transition. In the following year Braicovich and coworkers, after pioneering studies with the AXES spectrometer at the ESRF [6], indeed measured for the first time a single magnon dispersion with soft X-ray RIXS at the ADRESS beam-line at the Swiss Light Source, using the Cu L_3-edge of La_2CuO_4 in a measurement set-up with a combined energy resolution of 140 meV [7]. Fig. 2 shows that the measured RIXS magnon dispersion falls on top of the INS results for the same compound. This agreement between INS and RIXS is very satisfactory, but this comparison of the measured dispersions actually fails to highlight the most remarkable aspect of the RIXS experiment: whereas the INS data were collected on an array of seven mutually aligned single crystals with total mass 48.6 g., the RIXS data were collected on a merely 100 nm thick film of La_2CuO_4 . These experiments prove that magnetic RIXS on nanoscale structures is entirely possible.

Since then the field of magnetic RIXS on cuprates has flourished, going beyond the perhaps rather mundane insulating state of La_2CuO_4 towards for instance the doped cuprates which exhibit high temperature superconductivity. In these an intense paramagnon mode has been identified [7, 8], which had gone unnoticed in neutron scattering for many years (in INS its first fingerprints were uncovered by Lipscombe and coworkers only in 2009 [9]). This dispersing paramagnon mode is present in a series of different optimally doped high temperature superconductors and is believed to be of fundamental importance to the electronic pairing and hence the mechanism behind the superconductivity in these cuprates [8]. In determining magnetic RIXS cross-sections there were also considerable advances [10–13] and the field was furthered also to low dimensional cuprate quantum magnets such as spin-chains systems [14], spin-ladder materials [15] and various two-dimensional cuprates [16].

Whereas the potential of magnetic RIXS for other magnetic compounds consisting of 3d transition metal atoms is clearly enormous, due to the relative youth of the technique and the scarcity of beam-time experimental results are still sparse. In NiO purely magnetic excitations have been resolved [17], but their dispersion has not been mapped out yet. Very recently magnons have been observed in iron-pnictide superconductors [18], which appear to exhibit an intriguing paramagnon mode similar in character to the ones observed in high T_c cuprate superconductors [7–9].

Inspired by the success of soft X-ray magnetic RIXS at the copper L-edge at around 930 eV, the idea developed to explore the possibility for hard X-ray magnetic RIXS at the iridium L-edge [19], which for this 5d element is at around 11.2 keV. Due to their strong spin-orbit interaction of the 5d valence shell electrons in iridates [20], iridium-oxides exhibit a wealth of different magnetic phases, among which possible ones of non-trivial topological nature [21]. This year indeed the magnons of Sr_2IrO_4 were measured with hard X-ray magnetic RIXS [22] in an experiment at the Advanced Photon Source in Argonne where an instrumental energy resolution of 130 meV was reached, see Fig. 3. We believe also this marks the beginning of a new field, where magnetic RIXS



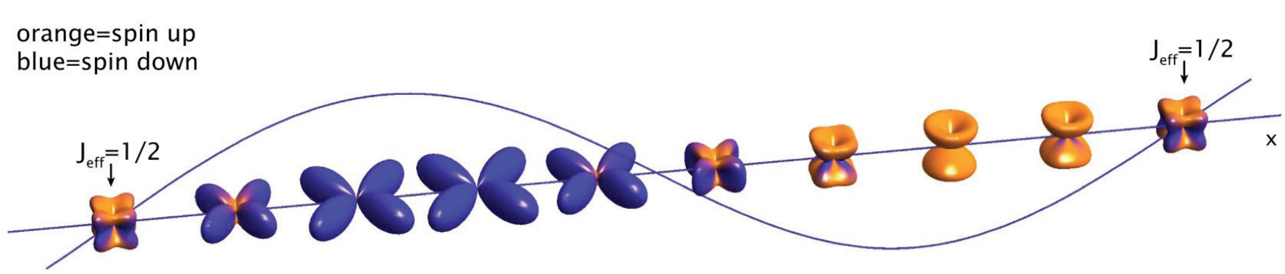


Figure 3: Top: Single magnon dispersion of Sr_2IrO_4 measured by magnetic RIXS at the iridium L_3-edge at 11.2 keV [22]. Bottom: real space representation of this magnetic quasiparticle which due to the very strong spin-orbit coupling in the Ir 5d-shell has a strongly mixed spin and orbital character.

can reveal the spin dynamics in 5d transition-metal systems, including also for instance osmates, a material class where INS is rather difficult because of the neutron-absorbing characteristics of the nuclei involved.

The width and depth of the advances in magnetic RIXS in the long term will depend on the potential for further improvements in energy resolution. Whereas it is obvious that for resolving magnetic excitations at the lowest energy scales of around 25 meV and below, inelastic neutron scattering on bulk materials will be unbeatable by the next few generations of RIXS instruments, for magnetic modes somewhat higher in energy, which are much more difficult to probe with INS, the situation is already quite different. For some material classes, in particular cuprates and iridates, RIXS is able to deliver results that are at present on par or even better than INS. However, a great advantage of magnetic RIXS is that it requires only tiny sample volumes – this year the magnetic excitations in a copper-oxide heterostructure of merely a single layer thick have been resolved [23] – so that for the first time the fundamental magnetic quasi-particles of nanostructures have become experimentally accessible. This is one of the basic considerations that motivates the push towards an energy resolution of 10 meV at the copper L-edge in the next generation soft X-ray magnetic RIXS beamlines of NSLS II at Brookhaven and the upgrade program of the ESRF. Similar advances in energy resolution are expected within the year for magnetic RIXS at the hard X-ray L-edges. All this, we believe, makes the prospect for RIXS to become another strength in the research of magnetism, in particular to study the fundamental spin-dynamics of magnets and to probe their magnetic quasi-particles, quite bright.

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