Journal of Magnetism and Magnetic Materials 400 (2016) 41–46

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

The promise of resonant inelastic X-ray scattering for modern Kondo physics

J.N. Hancock [a,*], I. Jarrige [b]

[a] Department of Physics and Institute for Materials Science, University of Connecticut, Storrs 06269, CT, USA
[b] National Synchrotron Light Source II, Brookhaven National Laboratory, Upton, NY 11973-5000, USA

ARTICLE INFO

Article history:
Received 25 June 2015
Received in revised form
3 August 2015
Accepted 5 August 2015
Available online 24 August 2015

keywords:
Kondo
RIXS
Moment screening

ABSTRACT

Developments in synchrotron accelerator and end station capabilities have brought new techniques to bear on complex matter. The recently enabled technique of resonant inelastic X-ray scattering (RIXS) has emerged as a serious contender of inelastic neutron scattering as a probe of magnetic structure in correlated electronic materials, but can also reveal perspectives on electronic structural not attainable by optical conductivity or angle-resolved photoemission. Here we introduce the basics of this powerful technique and discuss the prospects to turn this probe onto the charge excitations in Kondo lattice systems.

The study of the impurity Kondo problem in condensed matter has brought about many changes in physics, including the innovation of renormalization group formalism and which nucleated many notable contributions to theory of many body systems [1]. When the Kondo Hamiltonian is generalized once to include fluctuations of the valence state, and again to a periodic array of ‘impurity’ sites, one gets the periodic Anderson model (PAM), which contains a rich variety of electronic phases. Much of the physics of the heavy fermion class of materials is believed to be described by this electronic Hamiltonian, but many mysteries remain regarding the connection between theory and experiment, highlighting the importance of the development of new tools to probe complex correlated matter. Below, we contextualize the emerging technique of resonant inelastic X-ray scattering (RIXS) against related mainstay techniques and demonstrate its use to address the electronic structure of a particular material ($YbInCu_4$) in the physics of strongly correlated electron systems.

1. RIXS among other X-ray techniques

Core electrons are high energy bound electrons with filled-shell electronic configuration and wavefunctions which are extremely localized around the nucleus. For the pursuit of understanding materials behavior, these highly atomic electronic states are considered unimportant degrees of freedom because their excitations occur at a very high energy scale (>10 eV) compared to even the largest scales of valence electron excitations. X-ray interaction with materials can excite the deeply bound core electrons and promote them to unoccupied states. Across many scientific disciplines, X-ray spectroscopy is a mainstay of experimental research and characterization.

X-ray absorption (XAS) is a very important scientific tool, which can be regarded on many different levels. Often, it is used to simply provide a chemical fingerprint permitting estimates of stoichiometry, while in more advanced treatments, detailed analysis of the near-edge structure can be used to classify the valence states of particular atomic species in nonconducting materials [2,3]. Prominent examples of spin-off variants of direct XAS are extended X-ray absorption fine structure (EXAFS) [4,5], wherein the echoes of photo-excited electrons can be read from spectra to reconstruct the local environments of the resonant species, and X-ray magnetic linear/circular dichroism (XMLD/XMCD), wherein one senses the spin imbalance on a particular site through absorption of linear or chirally polarized photons [6]. Important work in the 1980s and 1990s [7,8] showed that this information was related in a simple way to the species-specific expectation value of orbital and spin magnetization components in the ground state, a development which catalyzed X-ray dichroisms rise to prominence as a materials characterization technique. Each of these probes are useful in their own right for the study of local structure and magnetic states in condensed matter.

When a core hole is photo-generated in a solid, this highly excited state awaits many possible fates that can be categorized into decay channels which eventually yield either a photon or a free electron (see Fig. 1). The most versatile and common-practice

* Corresponding author.
E-mail address: jason.hancock@uconn.edu (J.N. Hancock).

http://dx.doi.org/10.1016/j.jmmm.2015.08.017
0304-8853/

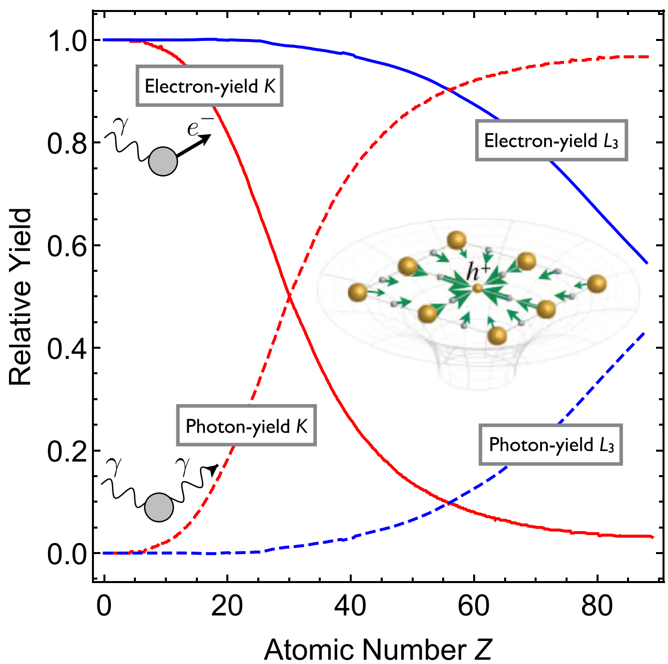

Fig. 1. Typical relative electron and photon yields resulting from X-ray excitation and for hole creation as a function of atomic number Z. For $Z > 30$, which describes most rare-earth and 5d electron systems, photon yield is significant. Inset: schematic showing the effects of core-hole creation on nearby valence electrons in a high-T_c superconductor structure.

method for collecting XAS data is through total fluorescence yield, where all photons emitted from the sample are collected without any special effort to discriminate their energies, polarization, or momenta. There are known conditions where direct absorption and total fluorescence yield give different results [45].

If the scattered photons are collected more selectively, using variable detection angle to segregate them by momentum, one has a powerful probe of electronic structure in real space with resonant elastic X-ray scattering (REXS) [9,10], also known as resonant diffraction. REXS is a fingerprint of static structures in matter complementary to neutron and nonresonant X-ray diffraction, which probes most directly magnetic and nuclear order. Because the REXS probe involves the highly atomic core levels, projections of atomic character of valence electron spin and orbital character are measured, giving a unique perspective in comparison to the more traditional techniques of neutron diffraction and nonresonant (Thomson) X-ray scattering.

If one discriminates the outgoing photons by energy as well as momentum, we have the developing technique of resonant inelastic X-ray scattering (RIXS). Differences in efficiency of this process for different kinematically distinct photon states carry direct information of the fundamental excitations of the material under study and is sensitive the fluctuations of charge and spin. The information carried by the photon field is very rich, and the ability to control and discriminate the photon degrees of freedom and prepare the state of the material in a well-controlled way is crucial to maximizing the return on the worlds RIXS investment.

2. Types of RIXS processes

RIXS differs from XAS in that it discriminates the emitted photons in the core-hole decay process by energy and often momentum and polarization. As with resonant Raman spectroscopy, one is then concerned with the ground state of physical interest, an intermediate state (which contains a core-hole at an X-ray edge), and a final state. RIXS can further be subdivided into the categories of high energy transfer (HET-RIXS), wherein the final state also contains a core hole, and low energy transfer (LET-RIXS), wherein the final state again involves the lower-energy ($<$10 eV) scales most relevant to material behavior.

HET-RIXS is often collected in a momentum-integrated fashion, as the process, highly atomic in nature, is not expected to have a strong momentum dependence. The kinds of information that one gleans regard the orbital occupancies of specific elements, and give particularly strong contrast in systems which feature valence instabilities [11–14]. The experimental requirements of this form of spectroscopy are not highly demanding, due to the high signal level, muted momentum dependence, and intrinsic broadening of the core-hole-containing final states.

LET-RIXS on the other hand ultimately generates low-energy excitations which are contained in the effective Hamiltonian relevant to the macroscopic low-energy behavior of physically interest. Far-reaching physical concepts arise from materials models in the spirit of the t-J, Hubbard, Holstein, periodic Anderson, and other minimal many-body Hamiltonians. As such, momentum dependence in LET-RIXS is not only expected, but constitutes a direct measurement of the dispersion of electronic and other collective excitations in the solid. During the recent period of rapid development, perspective on the character and dispersion of ligand orbital [15–17], crystal field [18,19], orbiton [20], magnon [21–24], bimagnon [24–26], paramagnon [27], polaron [28], Mott-gap [16,29], and even spin-2 triplon excitations [30] has been delivered by LET-RIXS. In cases, coupling between these degrees of freedom have been revealed. Because of the sensitivity to the unique combination of atomic species, valence state, orbital character, momentum, and energy, the information brought by RIXS probes is precisely what is needed to bridge the gap between atomic physics and the collective behavior of materials. As a burgeoning spectroscopic probe, the full expanse of the information one can gain has yet to be realized, but its value to the science community has become indisputable in recent years following provision of fresh perspectives on pointed questions in quantum materials long left open by the traditional techniques of inelastic neutron scattering (INS) and angle-resolved photoemission spectroscopy (ARPES) [11,27].

3. RIXS as a complement to ARPES and INS

A particularly profound gateway result of RIXS is the experimental discovery that a single spin flip can be produced by purely electromagnetic means (LET-RIXS). This effect is now well-understood [31], and single magnons have been observed using RIXS spectroscopy in 5d iridates [32] and 3d transition metals [21] with magnetic order. In cuprates, the magnetic signal has been tracked successfully to high doping levels, revealing that spin susceptibility exists in even strongly doped superconductors on the 300 meV scale [27]. An important theoretical element in the physics behind the scattering mechanism which makes these observations possible involves the extremely strong spin–orbit coupling of the 2p core level in a spin-polarized transition metal, which opens a photoelectron recombination pathway differing from the absorption pathway through the spin index [31].The importance of this discovery cannot be overstated, as it provides a method of studying magnetic excitations with numerous advantages over neutron scattering. Indeed many groups across the world, whose activities were traditionally based in neutron scattering, have recently embarked on successful RIXS activities. RIXS has clear advantages over inelastic neutron scattering: (i) clear information on which orbitals contain spin, (ii) high spatial resolution, and avenues to magnon microscopy, (iii) high signal can