Neutron and Photon Spectroscopy

General consideration on this lecture

This 3 hours lecture has the ambitious goal to give an overview of most spectroscopic techniques using neutron of light as probing particles. It has not the pretention neither to be exhaustive nor to describe all the possibilities and potential of each technique. It is a presentation of the basics of each technique, with simple pictures and description of the processes involved, with examples of information that can be extracted from it. This lecture is divided in two parts : Part I is an introduction of general concepts of inelastic process, followed by several experimental techniques excluding all that concerns magnetism. Part II is dedicated to magnetism, including some of the Part I experimental technique and new ones. There is many classifications possible for an overview of spectroscopic techniques. For both parts, I decided to separate them in two sections according to the physical process at play :

- inelastic scattering that can be viewed as one single photon/neutron that has its energy and momentum modified by interacting with the sample
- absorption spectroscopy that involves the annihilation of the probe, and its eventual consequences : emission/fluorescence.

Part I : introduction

Basic considerations on inelastic process
This part is dedicated to general consideration on inelastic physics, and different experimental spectroscopic techniques that do not involve directly magnetism. The first section describes the basics consequences of momentum, energy conservation law and their combination. I gave orders of magnitude for probes and most frequent excitations, concluding on the relation between experimental setups and their associated excitations measurable, underlying intrinsic limitations due to these basics concepts.

Scattering technique to probe lattice and charge excitations
The second section deals with inelastic scattering, consisting in considering a monochromatic incident wave (neutron of photon) interacting with a sample, resulting in a final monochromatic wave detected. I choose to describe inelastic neutron scattering (INS), inelastic X-ray scattering (IXS), and Raman scattering that all fulfill this description. For all of these techniques, I first derive the Fermi Golden Rule to describe the interaction, and understand the origin of the different terms in the scattering cross section. A description of those terms follows together with the specificities resulting from it. I complete this theoretical description by a presentation of standard experimental setup associated and few example of what can be measured. A small summary recalls the specificities of each technique, and the information it gives access to. In particular, INS and IXS are compared in one slide to highlight their differences despite their apparent similarities.

Photon-absorbing techniques : Spectroscopy
The third section is devoted to photon absorption processes. A description of Infrared (IR) Spectroscopy as a complementary technique to Raman is introduced theoretically, with the

very same examples used for Raman for a sake of clarity. A standard Fourier Transform IR Spectrometer is presented together with the mathematics to retrieve dielectric function from the reflectometry. Several examples of IR results both in reflection and transmission geometry are given with their references. A summary of IR technique characteristics and information it give access to ends this part.
IR Spectroscopy is followed by an introduction to the mechanism of X-ray absorption. After several example of core-hole creation and nomenclature associated, I describe few properties of absorption (element and oxidation state sensitivity, symmetry of the absorbing atom dependence etc…) and present few examples. Introducing XAS is actually mandatory to understand the RIXS technique that follow.
Indeed, RIXS process start with the creation of a core-hole as XAS, but involve a second photon process that is the emission (XES). I present then the different emission lines (fluorescence) and their nomenclature. I make then a distinction of direct and indirect RIXS. In direct RIXS, we simply consider the succession of the 2 processes : the intermediate state does not evolve between absorption and emission. It is sensitive to valence (XAS) and conduction (XES) states. In indirect RIXS the intermediate state interacts with the electronic bath and may involves electronic excitations. After a short description of the experimental technique of RIXS, I present several example of hard and soft RIXS results and terminate with a summary of key feature of this technique.

Part II : magnetism

Introduction to magnetic excitations

This second part is dedicated to magnetism and mainly concerns the measure of magnetic excitations. This is the reason I begin this part by listing the most common magnetic excitations such as magnon, spinon, crystal field and mixed excitations. I choose to present all excitations within the same framework and the same "local" picture. However, it is important to keep in mind that each sketch is only a local representation of the excitation (flip of the spin for example), but are actually delocalized spatially for energetic reason.

Scattering techniques probing magnetic excitations

One of the major technique to study magnetic excitation is certainly Inelastic Neutron Scattering (INS) as it enables to measure up to the dispersion of the magnons. I give an introduction of the cross section for magnetic scattering of neutrons, starting from the simple interaction of the neutron spin with spin or orbital contribution from the sample. Few emphasis are made about this cross section that needs to be highlighted : the magnetic form factor's strong q-dependence, the sensitivity to specific component of the spins (geometric factor) and the linear dependence of intensity. After few examples of results from INS, I highlighted key differences to discriminate between phonon and magnon from neutron scattering. To further enable such discrimination I present polarized neutron scattering technique, reinforcing the specific-component sensitivity of the technique. An example is given disentangling the nuclear and magnetic part of a hybrid excitation using polarized neutrons. A summary of key features to remember for magnetic INS terminates the section.
To complete the scattering techniques, I present the sensitivity of Raman Scattering to magnetic excitation. Unlike INS, Raman scattering is sensitive only to zone-center excitations. Depending on presence of Spin-Orbit coupling, Raman scattering is sensitive to magnon or bi-magnon. It can also detect spin-phonon coupling due to renormalization of phonon through

magnetic exchange. Examples of such phenomena from the literature are given, followed by the summary of key elements of magnetism seen by Raman.

X-ray absorbing technique probing magnetic ground state and excitations

The next technique (XMCD) is not dedicated to the measure of excitations but of the ground state. It can actually be seen as a particular case of X-ray Absorption Spectroscopy with circularly polarized photons. Measuring the difference of the absorption between the two helicity of polarization enable to evidence magnetism. Due to selection rules, only ferromagnetism is concerned. Furthermore, it is possible to disentangle the orbital and spin contributions to ferromagnetism.

Another magnetic ground-state sensitive technique presented here is X-ray Emission Spectroscopy. Considering specific emission lines (namely, K_beta and L_gamma), it is possible to measure the evolution of the amplitude of the magnetic moment carried by the absorbing atom. It is particularly interesting to evidence High-Spin-Low-Spin transition as given in example.

The last and more advanced and recent technique introduced in this lecture is the soft Resonant Inelastic X-ray Spectroscopy. This presentation is dedicated to explain both simple selection rules and from hand how RIXS process may be sensitive to bimagnon at the K-edge. At the L-edge we see that in presence of spin-orbit coupling, it is possible to have access to single magnons and spinons. Few recent examples are given with a description of the soft RIXS spectrometer.

All examples taken from the literature given in this lecture have their references given in each slide.

References :

Inelastic Neutron Scattering

- *Theory of Neutron Scattering from Condensed Matter Volume I: Nuclear Scattering & Volume II: Polarization Effects and Magnetic Scattering* Stephen W. Lovesey

Inelastic X-Ray Scattering

- *Introduction to High-Resolution Inelastic X-Ray Scattering* A.Q.R. Baron **http://arxiv.org/abs/1504.01098**

Raman Spectroscopy

- *Infrared and Raman Spectroscopy* Edited by Bernhard Schrader

Infrared Spectroscopy

- *Solid State Physics Part II Optical Properties of Solids* M.S. Dresselhaus

XAS, XES and XMCD

- *Core Level Spectroscopy* F. de Groot, A. Kotani

RIXS

- *Resonant inelastic x-ray scattering studies of elementary excitations* L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, J. van den Brink **Reviews of Modern Physics 83 (2011)**

- *Using RIXS to Uncover Elementary Charge and Spin Excitations* C. Jia, K. Wohlfeld, Y. Wang, B. Moritz, and T. P. Devereaux **Physical Review X 6, 021020 (2016)**
- *Magnetism and Synchrotron Radiation : Towards the Fourth Generation Light Sources* E. Beaurepaire, H. Bulou, L. Joly, F. Scheurer