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Nuclear resonant scattering of synchrotron radiation: Applications in magnetism of layered structures

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ABSTRACT

Nuclear resonant scattering of synchrotron radiation has become an established tool within condensed-matter research. Synchrotron radiation with its outstanding brilliance, transverse coherence and polarization has opened this field for many unique studies, for fundamental research in the field of light-matter interaction as well as for materials science. This applies in particular for the electronic and magnetic structure of very small sample volumes like micro- and nano-structures and samples under extreme conditions of temperature and pressure. This article is devoted to the application of the technique to nanomagnetic systems such as thin films and multilayers. After a basic introduction into the method, a number of our experiments are presented to illustrate how magnetic spin structures within such layer systems can be revealed.

1. Introduction

Atomic nuclei can be very sensitive probes of condensed matter properties. A number of spectroscopic techniques have been developed to probe condensed matter via nuclear hyperfine interactions, constituting the field of *nuclear condensed matter physics* [1,2]. A very prominent method in this field is Mössbauer spectroscopy that probes internal magnetic and electric fields in the sample by analysing the energetic hyperfine splitting of the nuclear levels via absorption of γ-radiation. Traditionally, this technique is applied in the energy domain where the radiation from a monochromatic radioactive source is Doppler tuned to measure the absorption spectrum around the nuclear resonance with nano-eV resolution. The hyperfine interaction of the nuclei in the sample typically lifts the degeneracy of the nuclear levels that manifests as a splitting and/or broadening of the absorption line(s).

Synchrotron radiation effectively allows one to perform the Fourier transform of Mössbauer spectroscopy from the energy domain into the time domain: the radiation is energetically broad (in contrast to the mono-energetic γ-radiation emitted by a radioactive source), and it comes in pulses with a duration of 50–100 ps (in contrast to the continuously emitting radioactive source). Thus, these pulses excite all hyperfine-split resonances in the sample at the same time. In the subsequent coherent decay the interference of the emitted waves leads to a temporal beat pattern, very similar to the temporal acoustic beats of slightly detuned tuning forks. From such a beat pattern the hyperfine interaction parameters of the nuclei in the sample can be deduced.

Within the natural linewidth of the Mössbauer transition, the brilliance of synchrotron radiation sources exceeds that of radioactive sources by several orders of magnitude. This was realized early by Ruby [3] and the first observation of the time-based analog of Mössbauer spectroscopy a.k.a. nuclear resonant scattering of synchrotron radiation was reported by Gerdau et al. [4] in Bragg scattering geometry and later by Hastings et al. [5] in forward scattering geometry. Since then, the method rapidly developed and is now available at many 3rd-generation synchrotron sources (ESRF, APS, Spring8, PETRA III) around the world, covering a broad range in all fields of the natural sciences [2,6].

2. Basic principles of nuclear resonant scattering

Due to the narrow nuclear resonance width in the range of neV–μeV, the scattering process takes place on time scales ranging from μs to ns. This allows for a discrimination of the resonantly scattered radiation from the non-resonant charge scattering and fluorescence that proceeds on time scales below 10^{-15} s. A synopsis of the method of time-resolved coherent nuclear resonant scattering is given in Fig. 1.

The transmission of X-rays through a slab of material that contains resonant nuclei can be described in a straightforward fashion via the energy-dependent index of refraction

$$A(z, \omega) = \exp[in(\omega)k_0 z]A_0.$$

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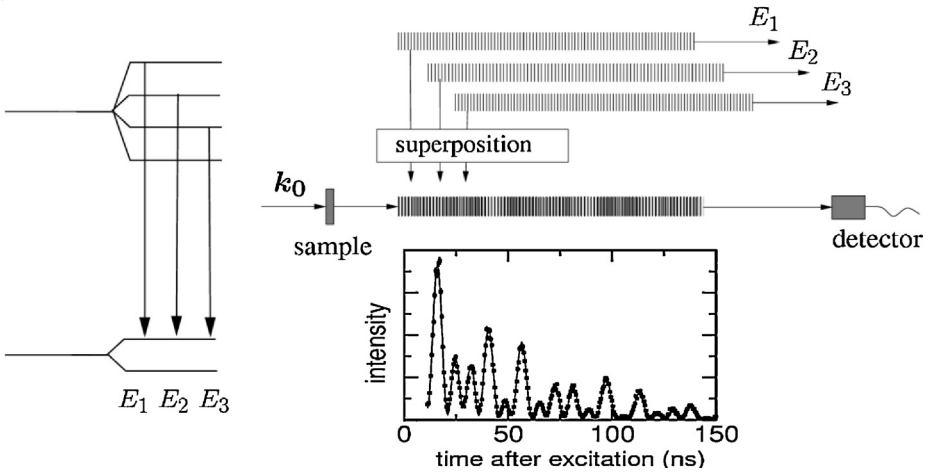

Fig. 1. Principle of coherent nuclear resonant scattering in forward direction. Transitions between nuclear levels are excited simultaneously by synchrotron radiation pulses at $t = 0$. In this example, three selected waves with slightly different frequencies are emitted from the decaying nuclei in the sample. Their superposition leads to quantum beats in the temporal evolution of the decay. This is illustrated here by overlaying three wavetrains with slightly different spatial frequencies, leading to a Moiré pattern. The intensity modulation of this pattern corresponds to the temporal quantum beats that are detected with a time-resolving detector like an avalanche photodiode (APD).

which gives the amplitude of the radiation field in depth z of the sample. A_0 is the amplitude of the incident radiation field. The index of refraction n is related to the forward scattering length M via

$$n(\omega) = 1 + \frac{2\pi}{k_0^2}\sum_k \rho_k M_k(\omega),$$

with p_k being the number density of the kth atomic species in the sample. These equations can be generalized to a multitude of scattering problems, ranging from simple forward scattering to anisotropic optics, thin-film reflection and Bragg diffraction from crystals and gratings [9]. The number N of open scattering channels determines the dimension of M, i.e., M is a scalar quantity for isotropic (polarization-independent) forward scattering, a 2×2 matrix for anisotropic (polarization-dependent) forward scattering, a 4×4 matrix for anisotropic (2-beam) Bragg diffraction which includes also nuclear resonant scattering in grazing incidence geometry. Correspondingly, the index of refraction $n(\omega)$ is a N-dimensional matrix.

Correspondingly, the quantities A_0 and $A(z)$ are vectors with N components that describe the amplitudes of the radiation field in the N scattering channels. This formalism naturally contains all multiple scattering processes within the sample, i.e., the resulting radiation field $A(z)$ is a self-consistent solution to the scattering problem within the dynamical theory of X-ray scattering.

The specific interaction of the photons with the atoms in the material is contained in the scattering length M. It is well known that optical properties of a system change drastically if the photon energy approaches an atomic resonance. This is valid for the complete spectral range from the infrared into the hard X-ray regime. The remarkable features of X-ray scattering from inner-shell resonances have been discovered and exploited when high-brilliance synchrotron radiation became available [10,11]. This applies for X-ray scattering from nuclear resonances as well [4,6,12].

Fig. 2 shows the real and imaginary parts of the forward scattering amplitude of ^{57}Fe over an energy range from 6 keV to 16 keV. Around 7 keV one observes the electronic K-edge of the material as it results from excitation of an electron in the K-shell into the continuum. At a photon energy of 14.4 keV one observes an extremely sharp feature that results from resonant excitation of the ^{57}Fe nucleus. The shape of this resonance becomes apparent only if one blows up the energy scale to the neV regime. The vertical axis is scaled in units of the classical electron radius r_0 which is the scattering length of a single electron. Thus it appears that near the resonance the scattering strength of the nucleus corresponds to an atom with $Z = 200$! This means that out of the small energy range around the resonance a very strong scattering signal can arise that exceeds that of electronic resonances by far [2].

Near the resonance the scattering amplitude of an atom with a single nuclear resonance can be written as

$$M(\omega) = E(\omega) + N(\omega),$$

where E and N are the electronic and nuclear contributions to the scattering length, respectively:

$$[E(\omega)]_{\mu\nu} = (\boldsymbol{\varepsilon}_\mu \cdot \boldsymbol{\varepsilon}_\nu)[-Zr_0 + f_e' + if_e''] = (\boldsymbol{\varepsilon}_\mu \cdot \boldsymbol{\varepsilon}_\nu)\left[-Zr_0 + f_e' + i\frac{k_0}{4\pi}\sigma(\omega)\right]$$

$$[N(\omega)]_{\mu\nu} = (\boldsymbol{\varepsilon}_\mu \cdot \boldsymbol{\varepsilon}_\nu)[f_n' + if_n''] = (\boldsymbol{\varepsilon}_\mu \cdot \boldsymbol{\varepsilon}_\nu)\frac{f_0}{x+i} = (\boldsymbol{\varepsilon}_\mu \cdot \boldsymbol{\varepsilon}_\nu)\left[\frac{f_0 x}{x^2+1} + i\frac{f_0}{x^2+1}\right]$$

where Z is the atomic number, r_0 the classical electron radius, and σ is the total absorption cross section. $(\boldsymbol{\varepsilon}_\mu, \boldsymbol{\varepsilon}_\nu)$ are the vectors of the polarization basis. Their scalar product that appears in the expressions above indicates that the scattering process conserves the polarization, i.e., the 2×2 matrices E and N are diagonal. This is in general not the case if the nucleus is subject to an electric or magnetic hyperfine interaction, as we shall see below.

The nuclear scattering length $N(\omega)$ is given here for a single, unsplit resonance line (i.e., no hyperfine interaction). $x = 2(E - E_0)/\Gamma_0$ denotes the deviation of the energy from the exact resonance energy E_0 measured in units of the natural linewidth Γ_0 of the transition. f_0 expresses the oscillator strength of the nuclear resonance:

$$f_0 = \frac{f_{LM}}{2k_0}\frac{2I_e+1}{2I_g+1}\frac{1}{1+\alpha}$$

where f_{LM} is the Lamb–Mössbauer factor that describes the relative fraction of photons that are elastically scattered (i.e., without recoil). I_e and I_g are the spins of the ground and excited nuclear state, respectively, and α is the coefficient of internal conversion.

For the 14.4 keV transition of ^{57}Fe we have $\alpha = 8.2$, $I_g = 1/2$, $I_e = 3/2$ and $k_0 = 7.3 \times 10^{10}\ \mathrm{m}^{-1}$. In α-Fe at room temperature we have