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Determination of hyperfine fields orientation in nuclear probe techniques

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ABSTRACT

One of the most popular nuclear probes, ^{57}Fe is used for the investigation of orientations of hyperfine fields and also for the determination of other important properties. In particular, the orientation of iron magnetic moments can be unambiguously determined, including its signs. Experiments with polarized radiation are presented with regard to selected systems. Orientation of electric field gradient is used for acquiring information about the shape of the texture-free spectra. Applications on the analysis of iron-based superconductors are presented.

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1. Introduction

Nuclear probe techniques offer an investigation of the hyperfine fields, resulting in an interaction of the nuclear spin and the electronic environment. These interactions cause shifts or splitting of nuclear levels. Electromagnetic transitions between the levels can be measured by Mössbauer spectroscopy (MS), γγ time-differential perturbed angular correlation (TDPAC), nuclear magnetic resonance (NMR), and many other techniques. Hyperfine fields are tensor quantities that cause anisotropy of the interacting radiation intensity. This, in turn, allows the determination of orientation of the local electric field gradient tensor and the direction of the hyperfine magnetic field.

Mössbauer spectroscopy is a well-established technique that is used for characterization of hyperfine interactions of the atom probes, including iron being one of the most important ones [1]. The relatively long lifetime of the excited ^{57}Fe nuclear level ($I_e = 3/2$) and recoilless processes of emission and absorption [2] result in a small natural width of the Mössbauer spectral lines, which results, in many cases, in the nuclear levels being well separated; that is, spectral lines do not overlap. The observation of separate transitions and the dependence of their intensity on the sample orientation are particularly important for unambiguous interpretation of the measured spectra. In many cases, this results in avoiding one of the serious problems frequently found in any kind of spectroscopy: correct separation of the overlapping spectra components.

In standard Mössbauer spectroscopy, nonpolarized, monochromatic radiation is used. Some information about the orientation of hyperfine interactions can be obtained by the use of nonpolarized radiation. However, a more complete insight is achieved by the use of different polarization states of photons. The aim of this article is to demonstrate the designing of the experimental setup for achieving monochromatic gamma radiation with the required polarization - circular or linear - and how to receive useful information about the orientation of hyperfine fields from the observed spectral line intensities. We will discuss how to use this information for the characterization of macroscopic physical properties. The examples are selected in such a manner that no advanced mathematical treatment is required and the interpretation of the results is straightforward.

2. Mössbauer Measurement With ^{57}Fe Isotope

Standard Mössbauer sources are commercially available, and they contain radioactive ^{57}Co isotope diffused into some metal matrix. The half-life of ^{57}Co is about 272 days. One of the possible states in the decay cascade is the excited ($I_e = 3/2$) ^{57}Fe nucleus, being a source of the 14.4 keV monochromatic nonpolarized radiation, used in Mössbauer spectroscopy.

The ^{57}Fe atom bounded in the crystal lattice or any other solid state may decay in a very specific way, without any transfer of energy to the elementary excitation (phonon). This process is called "recoilless

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emission." By moving the emitting source with controlled velocity, we may tune the energy of radiation because of the Doppler effect. At a certain velocity, the emitted photons may be absorbed in a recoilless process by a stationary sample containing ^{57}Fe in the ground state. The intensity of transmitted photons as a function of velocity results in the Mössbauer spectrum. Further details of the experimental technique can be found in refs. [1,2].

3. Dipolar Magnetic Interaction of ^{57}Fe Nucleus

The ^{57}Fe nucleus possesses a magnetic moment in the ground state (nuclear spin $I_g = 1/2$) as well as in the excited state (nuclear spin $I_e = 3/2$; see Fig. 1a). These moments interact with the hyperfine magnetic field acting on the nucleus (see vector ***B*** on the left-hand side of Fig. 1b). The interaction splits the $I_g = 1/2$ and $I_e = 3/2$ levels, resulting in sublevels being abbreviated by magnetic numbers $m_g = (-1/2, +1/2)$ and $m_e = (+3/2, +1/2, -1/2, -3/2$; see Fig. 1a). During transition of the nucleus from the excited to the ground state, the photon is being emitted in a certain direction, as shown by its wave vector ***k*** in Fig. 1b. All transitions have a well-defined polarization, depending on the direction of the orientation of the wave vector ***k*** and the hyperfine magnetic field ***B*** (see the left-hand side of Fig. 1b). The separation between the $I_g = 1/2$ sublevels (different m_g numbers) as well as between the $I_e = 3/2$ sublevels (different m_e numbers) is proportional to the hyperfine magnetic field ***B***, and it determines the absorption line positions of the Mössbauer absorption spectrum shown in Fig. 1c (the Zeeman sextet).

4. Resonant Filter

Standard Mössbauer sources are constructed by the diffusion of the radioactive ^{57}Co isotope into a high-quality matrix with a simple crystal structure, such as Cr, Pt, Rh, and CoO. Each matrix results in a specific isomer shift (see Fig. 2a).

By taking a monochromatic source with an appropriate isomer shift (Fig. 2a) and the magnetically split absorber (Fig. 1c), one can achieve

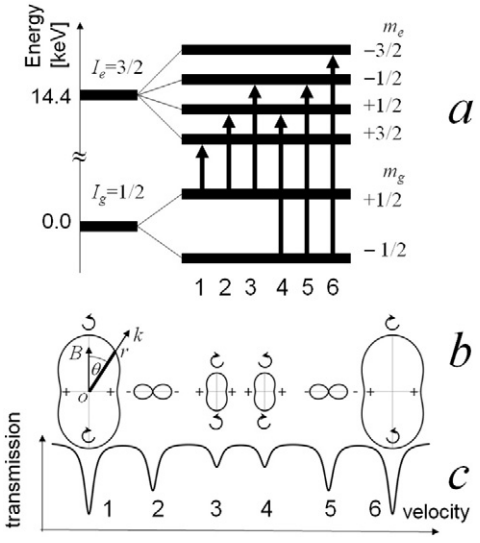

Fig. 1. a) Splitting of nuclear levels by hyperfine magnetic field. Allowed transitions are shown by arrows and abbreviated by integers 1, 2, … 6. b) Orientation of the hyperfine field ***B*** and the direction of the photon are shown on the left-hand side (***k*** is a wave vector of the photon). Polar plots show the intensity of absorbed photons for each transition. The intensity is proportional to the length *r*, shown schematically by a thick bar on the ***k*** vector. Absorbed photons are circularly polarized for $\theta = 0$ and $\theta = \pi$. Two opposite circular polarizations are shown by clockwise or counterclockwise arcs with arrows. The photons emitted perpendicularly to the hyperfine field ***B*** (angle θ between ***B*** and ***k***: $\theta = \pm\pi/2$) have a linear polarization. Two different linear polarizations are shown by *plus* or *minus* sign. c) Mössbauer spectrum of the magnetically split absorber (the Zeeman sextet). Absorption line numbers correspond to the transition numbers in a).

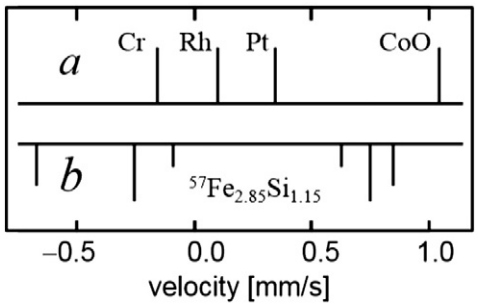

Fig. 2. a) Vertical bars showing position of emission line of ^{57}Co diffused into various matrices: Cr, Rh, Pt, and CoO. b) Vertical bars showing inner absorption lines (line nos. 3 and 4) of three Zeeman sextets present in the $^{57}Fe_{2.85}Si_{1.15}$ alloy.

absorption of photons at a zero Doppler velocity. This can be realized to some approximation by taking ^{57}Co in the Cr matrix (emission line located at −0.2 mm/s; see Fig. 2a) and $^{57}Fe_{2.85}Si_{1.15}$ ferromagnetic alloy (two absorption lines located at −0.25 mm/s and −0.1 mm/s; Fig. 2b). Therefore, the $Fe_{2.85}Si_{1.15}$ absorber can be attached to the radioactive source (as shown in Fig. 3), and the emitted photons will be absorbed in recoilless processes.

The absorption lines of $Fe_{2.85}Si_{1.15}$ located at negative velocities in Fig. 2b are the 3rd lines of the Zeeman sextets, and the transitions have the same polarizations (Fig. 1b, c). Complete spectra of Fe-Si alloys near Fe_3Si concentrations can be found in ref. [3]. The external magnetic field applied to the absorber (filter) results in orientation of the hyperfine magnetic field of iron in $^{57}Fe_{2.85}Si_{1.15}$ and, consequently, in well-defined polarization of the absorbed radiation (Fig. 3). Photons transmitted through the absorber (filter) will have opposite polarization. This is the main concept of polarization by the resonant filter technique [4,5,6]. The only difference in Fig. 3a and b is the direction of the external magnetic field acting on the $^{57}Fe_{2.85}Si_{1.15}$ filter and the polarization of the transmitted photons. Therefore, the magnetic field direction controls the polarization of the transmitted photons.

The whole constructions shown in Fig. 3a or b can be attached to the drive system and can then be used as a source of monochromatic, polarized radiation. The direction of the external magnetic field is realized by the set of permanent magnets [7,8,9]. A particularly important advantage of the proposed construction is the high chemical stability of the

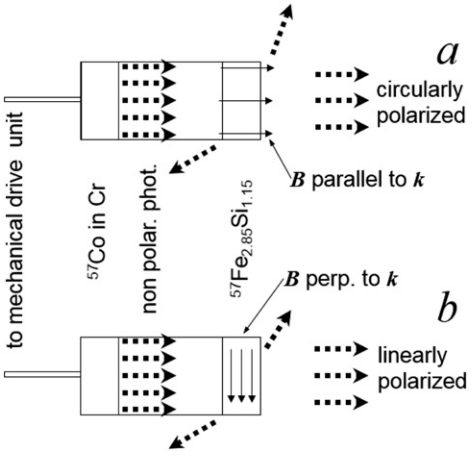

Fig. 3. a) $Fe_{2.85}Si_{1.15}$ absorber (filter) attached to the ^{57}Co source in the Cr matrix. Recoilless absorption occurs at a zero Doppler velocity. Hyperfine magnetic fields of the filter are oriented by the external magnetic field ***B***, resulting in transmitted photons that are circularly polarized. The external magnetic field ***B*** is parallel to the direction of the transmitted photons, i.e. the wave vector ***k***. b) The external magnetic field is oriented perpendicularly to the direction of the transmitted photons, resulting in their linear polarization. Photons are shown schematically by dotted arrows.