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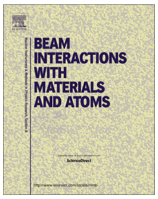

Mössbauer Magnetic Scan experiments

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ABSTRACT

We report an application of the Mössbauer Effect designed to retrieve specific information on the magnetic response of iron-containing materials. It consists in the measurement of the nuclear absorption of gamma-rays as a function of an external magnetic field for a specific nuclear transition between magnetically-split nuclear levels. The experiments, here termed Mössbauer Magnetic Scan experiments, were carried out recording the absorption of ^{57}Fe 14.4 keV gamma-ray in α-Fe at constant Doppler energies coincident with some of the spectral lines of the magnetically split Mössbauer spectrum. Due to the dependence of the transition probabilities on the relative orientation between the nuclear magnetic moment and the gamma-ray direction, the present application results in a useful method to study the magnetic-field evolution of the distribution of atomic-magnetic-moment orientations. The proposed technique inherit from the Mössbauer Spectroscopy the chemical-element selectiveness as well as the ability to differentiate responses from iron atoms located at inequivalent site or at different phases.

In this work, we show that the data analysis for these experiments depends on the sample thickness that the gamma-ray has to cross. For thin samples (i.e.samples with Mössbauer effective thicknesses lower than one) the magnetic-field dependence of the second-order-moment of the orientation distribution in the direction of the gamma ray is obtained. On the other hand, for thicker samples, although the data analysis is more complex, the dependences of the three second-order-moments of the orientation distribution are obtained.

The experiments were performed on two α-Fe foils of different Mössbauer effective thicknesses. They were chosen to represent the cases of thin and thick Mössbauer absorbers. The magnetic evolution of the orientations distribution is compared with results obtained from magnetometric measurements showing a good agreement as well indicating the complementarity of both techniques.

A complete description of the experimental set up and the formalism for Mössbauer Magnetic Scan data analysis are presented.

1. Introduction

Experimental methods measuring site-specific magnetic responses represent a basic and technological need in today's scientific world. The magnetic response of multiphase magnetic materials and of single-phase crystalline materials with nonequivalent magnetic atoms is of particular interest. This response either from different phases or from different structural sites of one particular phase of the materials to static and dynamic fields is a key point to understand the effect of local interactions in material properties. Single-phase nanomaterials, for instance, show that the magnetic response of spins in free surfaces and other interfaces is markedly different from that of spins in bulk regions. This is so due to the different atomic configurations and coordinations in the different locations.

Among others, some synchrotron techniques are selective to chemical elements and provide local information. X-ray Magnetic Circular Dichroism [1] has the property of being able to discern between the contributions to total magnetization of different atomic species present in a complex sample [2]. However, in cases in which the same element is located at non equivalent sites, for

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example Fe in magnetite, the dichroism signal is an average over these sites. Indeed, the problem on how the atomic magnetic-moments, located at octahedral and tetrahedral sites in this compound, individually respond to an applied magnetic field has been addressed by a more sophisticated synchrotron technique: site-specific diffraction anomalous near-edge structure [3]. This advantageous type of observation is not provided by most commonly used magnetometric techniques, which frequently give only information on the average magnetic moment component in one space direction. Therefore, the development of new, low cost, experimental methods and equipments with such ability is highly desirable.

Mössbauer Effect Spectroscopy is a widely used technique to study condensed matter [4,5]. It is a chemical-element selective technique and has the ability to distinguish probes in different phases or sites. Nowadays this spectroscopy is applied in a variety of disciplines [6], for example nano-science, biology, mineralogy, archeology, corrosion, solid state physics, to mention just a few. In the magnetic study of iron-containing alloys, this spectroscopy stands out due to its ability to fully characterize magnetic fields in internal domains. Besides, its high energy resolution allows to distinguish probes at inequivalent sites.

Another use of the Mössbauer effect comprises the measurement of gamma-ray resonant absorption as a function of some parameter (e.g. temperature, pressure, degree of advance of a reaction) at a constant Doppler energy. Detailed information about physical dependences on such parameters can be retrieved by this kind of experiments. This idea was used in several works where the energy was fixed at the centroid of the Mössbauer effect spectrum and the temperature was swept with the aim of determining characteristic temperatures (e.g. Curie temperature [7], crystallization on-set temperature [8], magnetic order temperature [8]). It was also applied to kinetic studies of phase transformations in which reaction rates were not slow enough to be accurately observed by Mössbauer spectroscopy. For example, selectiveness was essential to study recombination kinetics of the photodissociated Myglobin-CO [9] and nanocrystallization of $Fe_{73.5}Si_{13.5}Cu_1Nb_3B_9$ amorphous alloy [10]. More recently, we reported on some conceptual and practical advances of this approach accomplished by a set of experiments in which the Doppler energy was fixed at various values [11], or varied following a feedback [12] or a preset protocol, while the temperature was being changed. It was demonstrated that this method provides a higher degree of detail on material transformations than that obtained from the measurement of a limited number of conventional spectra.

Recently, the focus of our work has been extended to retrieve information of magnetic materials and their responses to applied magnetic fields. The methodology relies on the measurement of the resonant absorption of gamma-rays at fixed Doppler energies as a function of a magnetic field. In this case, a change in the relative intensities of the absorption lines is expected. This change is due to the nuclear-transition probability dependence on the angle between the nuclear magnetic-moment and the gamma-ray direction [4,5]. By selecting the Doppler energy coincident with one absorption maximum, the dependence of the magnetic-moment orientation on the applied magnetic field can be inferred.

In a previous work we have termed this kind of experiments as Mössbauer Magnetic Scan (MMS) experiments [13]. There, taking advantage of the tunability and selectivity of the technique, independent responses of the ^{57}Fe atomic magnetic-moments located at the nanocrystalline and amorphous phases of a nanocrystallized $Fe_{90}Zr_7B_3$ sample were observed. Distinctive features were found on each scan, according to the spectral line and structural phase to which they belonged. But it was not possible to explain the scans by using the thin absorber approximation formalism [14], not even qualitatively. Here, thin and thick mentions are related to the effective Mössbauer absorber thickness t, given by [14],

$$t = \sigma_0 f_A n d, \qquad (1)$$

where σ_0 is the maximum nuclear resonance absorption cross section, f_A is the recoil-free fraction, n the number of Mössbauer isotopes (in this case ^{57}Fe) per unit volume and d the sample thickness. Then, thin and thick absorbers correspond to effective thickness lower and greater than the unit, respectively. The main analysis difference between thin and thick absorbers, relies on the dependence of the spectral shape on the effective thickness. For thin absorbers, the absorption spectral function results linear in t, i.e. the line intensities and the spectral area are proportional to t, retaining the spectral shape. This linear behavior is not present in the case of thick absorbers [14–16]. As a consequence, thin absorber spectral analysis results more reliable. In fact, the linearity allows the decomposition of the spectrum in sum of sub-spectra and the quality of the information retrieved from the spectral line positions and their relative intensities does not depend on thickness accuracy.

Although the thin absorber formalism is widely used to analyze Mössbauer spectra even for samples with $t \approx 1$ [17], it does not account for changes in gamma-rays polarization as they travel through the sample. Indeed, for thick absorbers this fact usually cannot be ignored. Gonser et al. [16,18] observed significant changes in Mössbauer spectra in very thick samples (even remarkable changes as the apparition of additional absorption lines) that were evidently related with the polarization that the incident unpolarized beam acquires into the sample. Although for intermediate thickness the polarization effects are not reflected in the Mössbauer spectra line positions, this effect is clearly reflected in the spectral line intensities. Since MMS experiments focus on the line intensities, for a proper analysis, gamma-ray polarization process must be considered. To take into account these effects, we have extended the formalism introduced by Hirvonen et al. [19] and Daniels [20].

In this work, a thorough understanding of the MMS technique is accomplished. Their fundamentals and experimental details are described. Moreover, the mathematical procedure to properly analyze a given MMS curve is developed. The technique is tested with two α-Fe foils of 1 μm (thin) and 13 μm (thick), associated with effective Mössbauer thicknesses smaller and larger than one, respectively. It is confirmed that thick absorber MMS curves can only be analyzed under a formalism that jointly considers thickness and polarization effects. Besides, unlike the thin absorber case, information about the atomic magnetic-moment components orthogonal to the gamma-ray direction can be recovered.

2. Fundamentals

In this section, the underlying physics supporting the basic assumptions of MMS technique is described. The major relation that has to be discussed is the one between the direction of the atomic magnetic moment $\mathbf{m}$ and the spectral line intensities. Although there is an evident and direct relation between these intensities and the nuclear magnetic moment $\mathbf{m}_n$, the usefulness of the present application on condensed-matter-magnetism issue is more conditioned to a good understanding of the connection of spectral line intensities with atomic-magnetic-moment orientations. In this regard, a huge amount of interesting cases provides solid arguments holding a parallelism between the nuclear and the atomic magnetic moments. Here we put together these ideas and arguments because they are needed to interpretate the present methodology and also serve as a starting point to extend the interpretation to those cases in which this parallelism is not ensured.