Depth-Selective $^{57}$Fe Mössbauer Spectroscopy:
An Alternative

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Abstract. A method to obtain depth-selective $^{57}$Fe Mössbauer spectra is described. The depth selectivity is implemented by removing thin layers from a sample and measuring the conversion-electron Mössbauer spectrum. The layer spectra, i.e. the Mössbauer spectra associated with the removed layers, are then determined by calculating the number of electrons which originate from each layer and reach the surface of the sample. The computations are based on Liljequist's theory for $^{57}$Fe conversion-electron Mössbauer spectroscopy. In this way no energy-discriminating electron spectrometers are involved, and a simple proportional counter will suffice. The depth resolution is about 20–30 nm. The method is illustrated for an ion-implanted substituted YIG film, in which case the magnetization as a function of depth is obtained.

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In the past decade considerable progress has been made in obtaining depth-selective Mössbauer spectra (see e.g. [1–8]). It is now possible to discern the signal coming from the first 15–20 μm of the sample from that coming from deeper [1]. This is achieved by measuring the conversion-electron Mössbauer spectrum with an electron-energy discriminating detector, that is sensitive in the 7 keV region. In order to select the electrons coming from the top layer weight functions are used, which give the probability that an electron detected at some energy originates from a certain depth in the sample. By varying the electron-detector energy setting, electrons coming from various depths can be detected, but the depth resolution gradually deteriorates with the depth. Furthermore, since the transmission of the spectrometer is small in order to achieve a reasonable energy resolution, the samples have to contain a high concentration of the $^{57}$Fe isotope if reasonable signal-to-noise ratios are to be obtained in acceptable measuring times.

Recently Liljequist [1] has also developed a theory to compute the integral conversion-electron Mössbauer (CEM) spectra of a two layer system. In CEM the emitted electrons are detected with a large solid angle (2π radians) and with high efficiency. In the case that the system studied consists of a top layer and a bulk which possess different Mössbauer spectra Liljequist [1] showed that a layer thickness determination is possible with his theory. Vice versa, if the thickness of the top layer and the Mössbauer spectrum of the bulk are known, the Mössbauer spectrum of the top layer can be calculated. If several layers are present that have overlapping Mössbauer spectra or if there is a continuous change in spectrum parameters with depth, no straightforward solution is available. However, by repeatedly removing thin layers from the sample and subsequently measuring the electron Mössbauer spectrum, a set of spectra can be obtained from which it is possible to extract the spectra belonging to the removed layers. This procedure results in a depth-selective Mössbauer spectroscopy which takes considerably less time to perform than the energy discriminating approach. Also the depth resolution is independent of the depth itself.

This paper outlines the method and gives the equations necessary to obtain the layer spectra. A preliminary account of this approach was given in [10].

1. Determination of the Layer Spectra

In this section we arrive at expressions that relate a layer spectrum, i.e. the spectrum that belongs to a layer, to observed CEM spectra.
1.1. Model for a CEM Spectrum

In a CEM experiment one measures the number of electrons that emerge from the sample after a nuclear resonant absorption of a γ-quanta. In general, conversion and Auger electrons are detected, and secondary electrons, i.e., photo and Compton electrons. The number of electrons, \( S(v) \), that are emitted after nuclear resonant absorption and counted in a CEM experiment at a velocity \( v \) of the absorber with respect to the γ-ray source, can be written as

\[
S(v) = \int_0^d R(x) A(x, v) \, dx.
\]

Here \( d \) is the thickness of the absorber, \( A(x, v) \) is the number of nuclear de-excitations at depth \( x \) per unit of depth and \( R(x) \) is a response function of the absorber. \( R(x) \) gives the chance that an electron is detected at \( x = 0 \) due to a nuclear resonant absorption at depth \( x \). Since there are various electron emitting processes, \( R(x) \) is composed of a number of weight functions. A discussion of \( R(x) \) is given in the appendix.

1.2. Layer Spectra

We suppose the absorber to be composed of \( N-1 \) layers and the bulk \( N \). We assume that the number of nuclear de-excitations do not vary with depth within a layer. This implies that the reduction in the intensity of the γ-beam within a layer is neglected. The reduction in intensity after a layer has been passed, however, will be taken into account first order. The function \( A(x, v) \) is then reduced to a step function in the layers and (1) becomes

\[
S(v) = \sum_{i=1}^{N-1} A_i(v) \int_{x_{i-1}}^{x_i} R(x) \, dx + \int_{x_{N-1}}^{x_N} R(x) A_N(x, v) \, dx.
\]

The absorption by non-resonant processes of the recoilless γ-photons coming from the source are neglected in the layers, since these losses are, in general, at least one order of magnitude smaller. In the bulk all absorption processes are taken into account. The second term in (2) describes mainly the photo- and Compton electrons that are emitted from the layers, due to γ- and x-ray photons coming from the bulk.

The layered structure is defined physically by removing layers from the absorber, in our example by a chemical etch process. In a series of etch experiments a corresponding number of layers are removed at increasing depth. Before every etch step the CEM spectrum is measured, and thus \( N \) observed spectra are obtained.

For our system of \( N-1 \) layers and the bulk \( N \) before etching, the intensity of the source γ-beam after having passed through the \( i \)-th layer can be written as

\[
I_i(x_p, v) = I_0 e^{-\int_{x_{i-1}}^{x_i} \mu^B(x, v) \, dx}.
\]

After \( k-1 \) etch steps \((k \leq i)\) the number of de-excitations in the \( i \)-th layer has increased,

\[
A_i(v)(x_{i-1} - x_i)
\]

\[
= I_0 e^{-\int_{x_{i-1}}^{x_{i-1}} \mu^B(x, v) \, dx} \left(1 - e^{-\mu^B(x_{i-1} - x_{i-1})}\right).
\]

The CEM spectrum observed after \( k-1 \) etch experiments can then be written as

\[
S_k(v) = \sum_{i=k}^{N-1} A_i(v) \int_{x_{i-1}}^{x_i} R(x) \, dx + \int_{x_{N-1}}^{x_N} R(x) A_N(x, v) \, dx
\]

or, more compactly,

\[
S_k(v) = \sum_{i=k}^{N-1} A_i(v) U_{ki} + B_k(v)
\]

where

\[
U_{ki} = \int_{x_{i-1}}^{x_i} R(x) \, dx
\]

and

\[
B_k(v) = \int_{x_{N-1}}^{x_N} R(x) A_N(x, v) \, dx.
\]