Frequency Modulation of Resonant γ Rays in Iron Foils: Influence of the State of Magnetization*

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The properties of sidebands in Mössbauer spectra in the presence of a transversal radio-frequency field have been investigated in iron slabs. The relative intensity of the sidebands is analyzed in terms of fast phonon relaxation, because the single-modulation-index model does not account for the observed sideband intensities. The results obtained in the presence of the external magnetic field H_0 show that the generation of sidebands is caused by the oscillatory motion of the bulk magnetization rather than the domain-wall motion. The mean-square value of the atomic displacement, x^2, as a function of H_0 shows an abrupt decrease near the anisotropy field H_A in the hard direction (375 Oe) and drops to zero very rapidly. We point out that, concerning magnetostriction as a possible mechanism, the application of the formula $\Delta l = l_0 \Lambda_s (\cos^2\theta - \frac{1}{3})$ is correct only for powdered samples where the displacement of atoms is expected to be a quasistatic coherent phenomenon. However, in the case of thin slabs, where the wavelength of ultrasonic phonons at ω_{rf} is much smaller than the sample size, the application of the above formula does not account for the observed modulation. We have calculated x^2 by evaluating the average elastic energy density of the sample under rf irradiation. The strong modulation observed implies that the magneto-elastic coupling coefficient is much larger in the dynamic condition than in the static one. From our discussion it follows that magnetostriction could also be responsible for the published results on a low-magnetostriction material such as supermalloy.

SEVERAL recent experiments have established that the γ-resonant lines of a ferromagnetic slab in the presence of a radio-frequency (rf) magnetic field exhibit sidebands[1] strikingly similar to the sidebands generated by ultrasound Doppler modulation of the γ-ray energy.[2,3] Along the line of previous experiments,[1] we report here the results obtained on iron slabs of various thicknesses as a function of both the intensities of the rf field (H_{rf}) and the static external magnetic field (H_0). The new information thus obtained also enables us to understand better the phenomenology presented by other authors, and to establish a limit to the validity of their interpretations.

The analysis of the intensity of the sidebands in terms of a single modulation index is not consistent with the data, whereas agreement is found in all cases if such analysis is carried out on the basis of the quantum-mechanical treatment.[4]

Another feature, in the light of the experiments of Perlow,[1] is the appearance of sidebands at 4 Mc/sec in iron slabs. This result, and considerations on the magnetic anisotropy, makes the model of hyperfine field random flipping inapplicable, both for iron and for supermalloy slabs.

Furthermore, the results with external static fields H_0 show the disappearance of the sidebands for sufficiently large H_0 and shed new light on the role of magnetic anisotropy. The motion of domain walls is ruled out as a possible explanation. A semiquantitative calculation of the relative intensity of the sidebands is presented for the model of the magnetostriction.

If an enriched ^{57}Fe slab is irradiated with 4-Mc/sec magnetic field perpendicularly to the direction of propagation of the 14.4-keV γ ray, the well-known Mössbauer spectrum exhibits, at least for small H_{rf}, clear sidebands (Fig. 1.) As H_{rf} is further increased, the overlap of all the sidebands with the original spectrum degenerates into a broad double peak, very similar to those obtained by Perlow on supermalloy samples. A

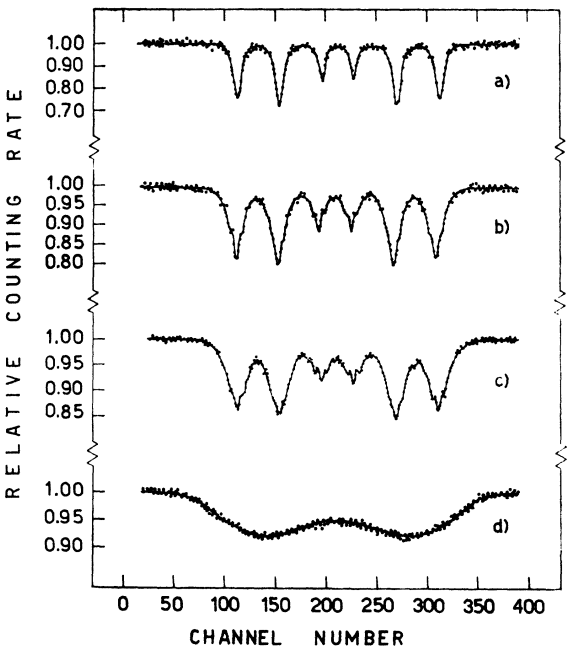

FIG. 1. Enriched ^{57}Fe foil (2.5 μ thick) irradiated with 4-Mc/sec magnetic field: (a) $H_{rf}=0$ Oe; (b) $H_{rf}=0.3$ Oe; (c) $H_{rf}=0.45$ Oe; (d) $H_{rf}=0.9$ Oe.

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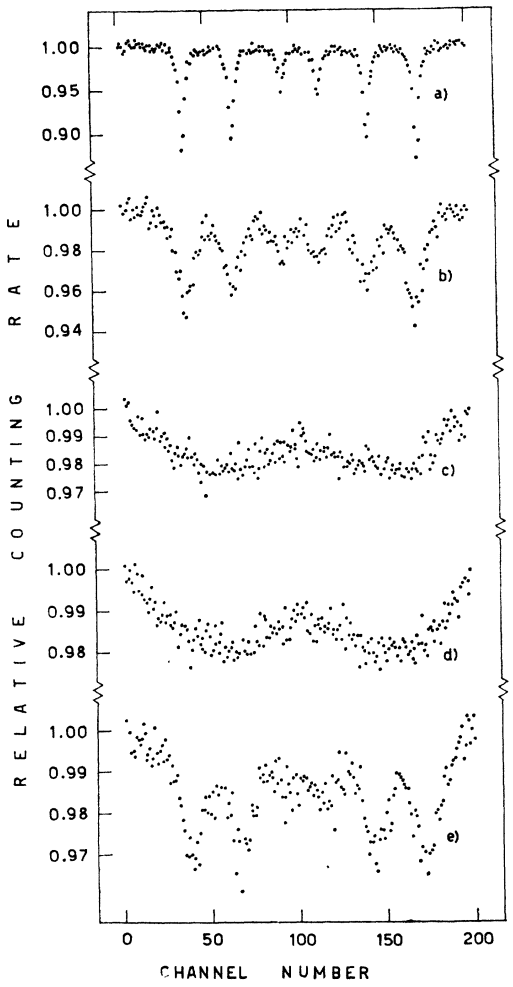

FIG. 2. Natural iron foil (12 μ thick) irradiated with 4-Mc/sec magnetic field: (a) $H_{rf}=0$ Oe, $H_0=0$ Oe; (b) $H_{rf}=1$ Oe, $H_0=0$ Oe; (c) $H_{rf}=1$ Oe, $H_0(\perp H_{rf})=30$ Oe; (d) $H_{rf}=1$ Oe, $H_0(\perp H_{rf})=300$ Oe; (e) $H_{rf}=1$ Oe, $H_0(\| H_{rf})=50$ Oe.

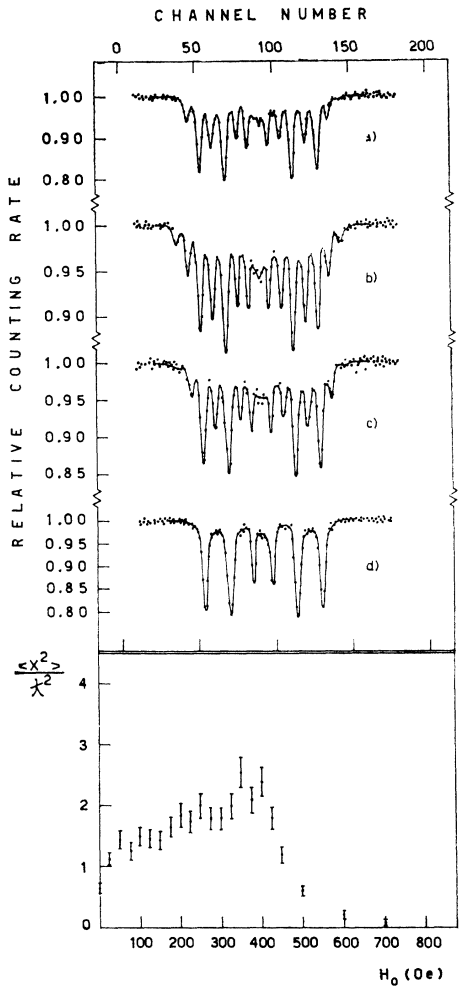

FIG. 3. Enriched ^{57}Fe foil (2.5 μ thick) irradiated with 13-Mc/sec magnetic field as a function of the external static field H_0, perpendicular to H_{rf}: (a) $H_0=0$ Oe; (b) $H_0=300$ Oe; (c) $H_0=500$ Oe; (d) $H_0=1000$ Oe. The argument of the Bessel function, x^2/λ^2, is plotted as a function of H_0 at the bottom.

static magnetic field H_0 perpendicular to both H_{rf} and to the direction of the γ rays enhances this effect, whereas H_0 parallel to H_{rf} quenches it partially (Fig. 2). The intensity of H_0 in this experiment was varied from 0 to 300 Oe.

A better insight on the influence of the external field H_0 can be gained by choosing a suitable position of the sidebands relative to the original Zeeman sextet, i.e., with a suitable ω_{rf}. In the case of iron, a convenient value for ω_{rf} is 13 Mc/sec. The external field H_0 was perpendicular to H_{rf} and measurements were performed every 25 Oe from 0 to 1000 Oe. Each value of H_0 was reached from above saturation: 5000 Oe. Some of the measured Mössbauer spectra are shown in Fig. 3.

A careful analysis of the intensity of the sidebands for each peak of the Zeeman sextet yields the following information: The sideband spectrum is described by the expression obtained by Abragam[4] following a quantum-mechanical treatment,

$$\tilde{W}(E)=\exp\left(-\frac{x^2}{\lambda^2}\right)\sum I_n\left(\frac{x^2}{\lambda^2}\right)W(E-n\omega_{rf}), \quad (1)$$

where $W(E)$ is the spectrum in the absence of rf field, x^2 is the mean-square value of the amplitude of atomic vibrations, and λ is the reduced wavelength of the γ radiation. The relative intensity of the sideband of order n is given by

$$\exp(-x^2/\lambda^2)I_n(x^2/\lambda^2),$$

where I_n is the Bessel function of order n. Abragam shows that the expression (1) coincides with the classical one, provided one introduces a distribution of "modulation indexes" m. This correspondence has been verified by other authors[3] by means of ultrasound Doppler modulation of the γ-ray energy. Only for very small

values of m is it possible to approximate the distribution of m's with a single value m, with the consequence that the intensity of the nth sideband is proportional to $J_n^2(m)$. All of our results and the data presently available in the literature are described by Eq. (1), although there are no examples of sideband spectra which can be fitted with a single modulation index m.

From the measured spectra, we have deduced the argument x^2/λ^2 of the Bessel function $I_n(x^2/\lambda^2)$. If we plot x^2/λ^2 as a function of H_0, we obtain the curve of Fig. 3. We note a sharp increase of x^2/λ^2 in the low-H_0 region (<50 Oe) where the mechanism of magnetization is dominated by the domain-wall displacement. A further increase occurs up to fields as high as 400 Oe, which is approximately the value of the anisotropy field. For larger values of H_0, x^2/λ^2 decreases markedly. The behavior of x^2/λ^2 as a function of H_0 strongly suggests that the generation of sidebands is caused by the oscillatory motion of the bulk magnetization M rather than the domain-wall motion. Domain walls have practically disappeared in the H_0 region where x^2/λ^2 is still increasing. In this region (100–400 Oe) the competition between the applied field H_0 and the anisotropy field H_A, averaged over the crystallites of the sample, results in a larger amplitude of oscillation of $\mathbf{M}$ under the influence of the rf field. As H_0 overcomes H_A in the hard direction (375 Oe), the magnetization is more and more inhibited in its motion by the net resulting field ($\mathbf{H}=\mathbf{H}_0+\mathbf{H}_A$), and correspondingly x^2/λ^2 drops to very small values.

As far as magnetostriction is concerned as a possible mechanism responsible for the sidebands, we can calculate the expected relative intensity of the effect in the following cases:

(i) Iron-powdered absorbers. If the mean diameter of the grains is smaller than the phonon wavelength at ω_{rf}, we expect that the average displacement of the atoms due to the magnetostriction is the result of a quasistatic coherent phenomenon. The displacement is therefore given by

$$\Delta l = l_0 \Lambda_s (\cos^2\theta - \tfrac{1}{3}), \tag{2}$$

which commonly represents the static magnetostriction in an isotropic medium, such as polycrystalline iron. Here Λ_s is the average magnetostriction constant ($=-7\times10^{-6}$ for iron),[5] and $\cos\theta = M/M_s$, where $\mathbf{M}=\mathbf{M}_0+\mathbf{M}_{rf}$, $\mathbf{M}_0$ and $\mathbf{M}_{rf}$ being the static and the rf components, respectively, of the magnetization $\mathbf{M}$. In the low-H_{rf} limit, we can calculate the modulation index

$$m = \Delta l \omega_\gamma / c = \tfrac{3}{2}\Lambda_s l_0 (\omega_\gamma/c)(\cos^2\theta - \tfrac{1}{3}).$$

The term in the last set of parenthesis in Eq. (2), for $M_0 \gg M_{rf}$, becomes

$$\cos^2\theta - \tfrac{1}{3} \approx 2(M_0/M_s)(M_{rf}/M_s).$$

Because $(M_0/M_s)M_{rf}/M_s \leq \mu_r H_{rf}/M_s$, where μ_r is the initial permeability based on the rotation magnetization, which for iron is approximately 29,[6] an upper limit for m is given by

$$m \leq \tfrac{3}{2}\Lambda_s (l\omega_\gamma/c) 2\mu_r H_{rf}/M_s. \tag{3}$$

By putting in Eq. (3) $\omega_\gamma/c = 7.28\times10^8$ cm^{-1}, $\Lambda_s = -7\times10^{-6}$, $|H_{rf}| = 2$ Oe, $l = 3\times10^{-4}$ cm, and $M_s = 21\,500$ G, we obtain, for 6-μ average diameter iron powder, a modulation index $m \leq 1.2\times10^{-2}$. In agreement with the experimental results on iron powders, such a small modulation index does not produce any detectable sideband pattern.

(ii) Iron-slab absorbers. In the present experiment, however, we are dealing with slabs of $\sim$1-cm diameter. In this condition the wavelength of the elastic waves generated by magnetostriction is smaller than the sample size. The situation is therefore not a static one and the application of Eq. (2) is not appropriate. That this is so is evident from the discrepancies between the observed intensity of the sidebands and the values of Δl thus calculated, which are $\Delta l \simeq 0.54\times10^{-2}\lambda$. In the thin-slab sample, the situation can be visualized as an incoherent distribution of elastic waves at $\omega = \omega_{rf}$ and the parameter of interest for the observed Doppler effect is therefore the mean-square displacement of each atom: x^2. If n_{rf} is the average population of phonons of frequency ω_{rf}, x^2 will be given by[4]

$$x^2 = (\hbar/3Nm)(\bar{n}_n + \tfrac{1}{2})/\omega_{rf}. \tag{4}$$

$\bar{n}_{rf}$ can be calculated if the average elastic energy density stored in the volume of the sample, $\langle E_{el}\rangle$, is known. $\langle E_{el}\rangle$ is a fraction k^2 of the average magnetic energy density $\langle E_m\rangle$. $\bar{n}_{rf}$ is given by $k^2\langle E_m\rangle/\hbar\omega_{rf}$. The calculation of $\langle E_m\rangle$ is performed in the state of remanence, where it is given by $(1/8\pi)\mu_r(H_{rf,eff})^2$, $H_{rf,eff}$ being the effective value of H_{rf}. We obtain thus $\langle E_m\rangle = 2.3$ erg/cm^3 and therefore $\bar{n}_{rf} = 2.9\times10^{19}k^2$ cm^{-3}. By substituting this value in Eq. (4), we obtain $x^2 = 8.7k^2\lambda^2$.

The values of x^2/λ^2 obtained in the present experiment indicate that the magneto-elastic coupling coefficient k^2 should be as large as approximately 10%. On the other hand, experimental results on the static "ΔE effect" in iron[7] suggest that the value of k^2 is less than 1%.[8] It must be concluded that either in dynamical conditions the transfer of magnetic energy into elastic energy is larger than in static conditions, or the model of the magnetostriction is not responsible for the sideband effect.

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[5] A. H. Morrish, *Physical Principles of Magnetism* (John Wiley & Sons, Inc., New York, 1965), p. 325.

[6] S. Chikazumi, *Physics of Magnetism* (John Wiley & Sons, Inc., New York, 1969), p. 263.

[7] R. N. Bozorth, *Ferromagnetism* (D. Van Nostrand, Inc., New York, 1951), p. 687.

[8] Note that $k^2 = d^2/S^H\mu_r$: see *Solid State Magnetic and Dielectric Devices*, edited by H. W. Katz (John Wiley & Sons, Inc., New York, 1950). For iron, $d = 0.7\times10^{-6}$ Oe^{-1}, $S^H = 0.88\times10^{-12}$ cm^2/dyn, $\mu_r = 29$, and thus $k^2 = 2\times10^{-2}$.

On the basis of our experiments, we cannot interpret the results with the model of the random flipping of the hyperfine field.[1] (The sidebands observed at 4 Mc/sec could not possibly be detected in supermalloy samples, as in Perlow's experiment, because of the large width of the Mössbauer lines at $H_{rf}=0$.) Furthermore, the calculation of x^2 given above for the case of iron has been carried out also for supermalloy samples. The relatively smaller values of k^2 for the latter can be compensated by the relatively larger magnetic energy density. Consequently, the model of the magnetostriction might be valid for supermalloy, if it is for iron.

In conclusion, the present experiments yield the following information:

(a) The sidebands observed in iron slabs of various thicknesses in the presence of a rf magnetic field cannot be described by a single "modulation index"; a quantum-mechanical derivation of their spectrum accounts satisfactorily for the obtained results.

(b) The dependence of the intensity of the sidebands on the static magnetic field H_0, perpendicular to H_{rf}, suggests that the domain-wall contribution, if any, to the observed effect is much smaller than the contribution of the rotation of domain magnetization.

(c) The magnetostriction model accounts for the intensity of the sidebands, if the magneto-elastic coupling coefficient k^2 is assumed to be much larger in the dynamic condition than in the static condition.

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First-Order Phase Transition in Europium Metal

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We have used Mössbauer-effect measurements of the hyperfine (hf) interaction in Eu metal to study the behavior of its sublattice magnetization in the vicinity of the magnetic ordering temperature. At 88.6°K, the hf field falls from 0.4 of the saturation value to zero. This results from the existence of a first-order transition coincident with the magnetic ordering. The possible causes of the transition are discussed. The temperature dependence of the hyperfine field just below the transition is analyzed in terms of critical-point theory. The results of thermal-expansion measurements are also presented and discussed.

I. INTRODUCTION

THERE has been a renewed interest in the behavior of substances in the vicinity of critical points of their phase diagrams[1] ever since the derivation of the "scaling laws." In particular, many investigations of magnetic systems in the vicinity of the ordering temperature have been performed, though there are a number of difficulties connected with the extraction of the relevant parameters from the experimental data. In particular, the temperature dependence of the sublattice magnetization has been measured for a number of compounds in the vicinity of the critical temperature. With this aim in mind, a measurement of the temperature dependence of the hyperfine (hf) fields in Eu metal was undertaken.[2] As a result of these measurements, we found that the paramagnetic to antiferromagnetic transition in Eu metal is of first order. This article mainly presents the experimental evidence of this and discusses the causes for the first-order transition, and only secondarily will we discuss the result in terms of a crtical-point analysis. For clarity, we have chosen to call the temperature at which the hf field disappears T_t ($=88.6\pm0.3$°K) in our sample. The term T_N represents the temperature at which the hf field, extrapolated on the basis of a power law from points just below T_t, would go to zero if the first-order transition did not take place. T_N is about 1°K above T_t.

In addition to the Mössbauer measurements, a thermal-expansion measurement was performed in order to check whether there was any anomalous change of the lattice parameters at the magnetic ordering temperature.

There are two previous measurements on the Mössbauer effect in Eu metal.[3,4] These experiments did not observe the sharp transition reported here, probably because of inadequate sample purity.

II. PROPERTIES OF Eu METAL

Eu is divalent in Eu metal, which has bcc structure. This can be inferred from the isomer shift observed in

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