Mössbauer observation of radio-frequency striction of the paraprocess in invar alloys

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(Submitted November 26, 1973)
Zh. Eksp. Teor. Fiz. **66**, 1777–1782 (May 1974)

Sidebands induced by radio-frequency (rf) pumping at 40 MHz with amplitude up to 10 Oe are observed in the Mössbauer spectra of Fe alloys containing 35 and 30 at.% Ni. Experiments carried out in external magnetic fields and pulsed rf fields show that the sidebands are caused by paraprocess striction. The results are discussed and estimates of the rf striction are presented.

1. The study of rf excitations in solids by using Mössbauer spectroscopy[1-5] is based on detection of the ultrasonic vibrations of nuclei. If the frequency of these vibrations ν is larger than the Mössbauer line width, then satellite lines (sidebands) separated from the parent line by $\pm n\nu$ occur in the spectra. As shown in[4], the main mechanism responsible for the generation of internal acoustic vibrations in magnetic materials is magnetostriction. In this model, the elastic ultrasonic vibrations of the nuclei are caused by rf oscillations of the sample magnetization.

Until now the rf fields used in Mössbauer studies of acoustic waves in magnetic materials were obtained by technical magnetization. The ultrasonic vibrations were generated by periodic changes in the linear magnetostriction due to boundary displacements or rotation of the magnetization vector. In the present work the ultrasonic vibrations of the nuclei due to a volume paraprocess striction were observed for the first time. This paraprocess striction accompanies the so-called true magnetization, and is determined by dependence of the exchange integral on the interatomic separation.

2. The Mössbauer spectra were obtained with a constant acceleration spectrometer. We used a 256-channel pulse analyzer. The sample was placed in the center of a flat coil formed by 10 turns of silver-plated copper wire. The sample served as the absorber in a Mössbauer transmission experiment. The coil was capacitance-coupled to the output of a 30-watt radio-frequency oscillator. The spectra were taken at (40.7 ± .8) MHz, the peak amplitude of the rf field (h_{rf}) being up to 10 Oe. The experiments were carried out with both continuous and pulsed-mode rf pumping. In the latter case the measurements were arranged in such a way that the odd channels of the analyzer displayed the spectrum in the absence of the rf power, and the even channels worked in its presence[6]. For this purpose the rf oscillator was synchronized by the pulses from the first trigger of the analyzer's address register. The duration of the rf pulse packet was 500 μsec, and the duty factor was equal to one half.

A single-channel amplitude discriminator was built to display the spectra in both even and odd channels on the oscilloscope screen. Its switching circuit allowed us to add background noise at the end of the experiment only into the odd channels of the analyzer, thus creating an offset separating the even- and odd-channel spectra.

The external static magnetic fields H_0 were produced by permanent magnets and were monitored with Hall probes.

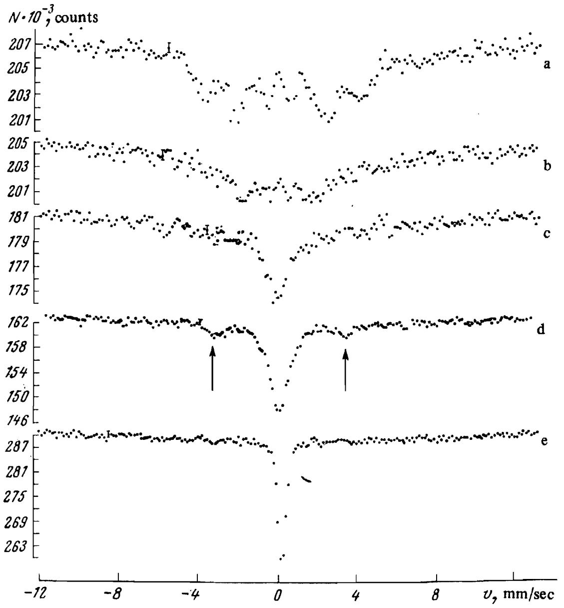

FIG. 1. Mössbauer spectra of the alloy Fe + 35% Ni with continuous mode rf pumping: a) h_{rf} = 0, b) h_{rf} = 3 Oe, c) h_{rf} = 4 Oe, d) h_{rf} = 5 Oe, e) h_{rf} = 10 Oe. The arrows in Fig. 1, d point to the satellite lines.

We studied foils of invar alloys Fe + 35 at.% Ni, and Fe + 30 at.% Ni. The foils were 25 μm thick; they had been annealed in hydrogen at 1000°C for 5 h to remove inhomogeneities, and were then slowly cooled in the furnace.

3. The Fe–Ni invar alloys were chosen because of the large value of their volume paraprocess striction. We should also emphasize that near the Curie temperature the paraprocess is the determining factor in magnetization, and is already apparent at weak fields[7].

Figure 1 shows the Mössbauer spectra of a 35% Ni sample at varying powers of the applied rf field in the continuous pumping mode. It can seem that the lines of magnetic hyperfine structure "collapse" as the power of rf field increases. This is due to an increase of the sample temperature, and results finally in a single narrow line corresponding to $T > T_c = 240°C$. However, in a certain power range, which corresponds to $T \le T_c$, satellite lines appear in the spectrum apart from the single broadened line. An example of such a spectrum is displayed in Fig. 1d. Since these satellite lines disappear at T_c, it is felt that they result from magnetic ordering in the sample.

The spectra displayed in Fig. 2 contain experimental

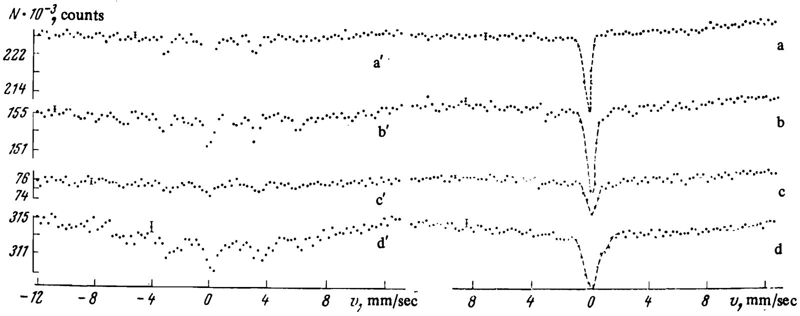

FIG. 2. Mössbauer spectra of the alloy Fe + 35% Ni with pulsed-mode rf pumping, $h_{rf} = 5$ Oe, in varying external magnetic fields $H_{0\perp} H_{rf}$. Spectra a), b), c), and d) were taken without rf power (even channels); a'), b'), c'), and d') - in the presence of rf power (odd channels); a) and a') $H_{0\perp}$ - 3μ Oe, b) and b') $H_{0\perp}$ - 120 Oe, c) and c') $H_{0\perp}$ - 190 Oe, d) and d') $H_{0\perp}$ = 470 Oe.

evidences that the observed satellites are associated with paraprocess striction. The figure shows spectra taken in the pulsed pumping mode. The odd-channel spectra (without rf field) are displayed to the right, while the even-channel spectra (in the presence of rf field) are on the left side of the picture. All the spectra were taken in external fields $H_0 \perp H_{rf}(H_{0\perp})$. The widths of the single lines (Fig. 2, a—d) are seen to increase appreciably as H_0 increases from 34 Oe to 470 Oe. This corresponds to an increase of 7 ± 1 kOe in the effective magnetic field H_{eff} felt by the nucleus. Such a change of H_{eff} in response to $H_0 \approx 0.5$ kOe can be explained only as H_{eff} being induced by an increase of the alloy magnetization in the paraprocess region near T_c[8].

Examination of the even-channel spectra (Fig. 2, a'—d') shows that the relative intensivity (I_{sb}) of the sidebands increases markedly with $H_{0\perp}$. I_{sb} is obviously proportional to the amplitude of $d\omega/dt$, where ω is the volume striction of the paraprocess $\omega = k[\sigma(T, H)]^2$; σ is the total magnetization referred to 0°K; k is a coefficient proportional to dJ/dr (J being the exchange integral and r the interatomic separation). Then

$$I_{sb} \sim k\sigma \frac{d\sigma}{dt}, \quad (1)$$

$$\frac{d\sigma}{dt} = \frac{d\sigma}{dH_{rf}} \frac{dH_{rf}}{dt} \sim \chi_{rf} h_{rf},$$

where $H_{rf} = h_{rf} \sin \nu t$ is the intensity of the rf field and χ_{rf} is the radio-frequency susceptibility. Finally, we get

$$I_{sb} \sim k\chi_{rf}\, \sigma h_{rf}. \quad (2)$$

From (2) it follows that I_{sb} increases with H_0, since in the paraprocess an increase of H_0 involves a sharp rise of the magnetization σ. On the other hand, χ_{rf} is maximal for the $H_{0\perp}$ fields. Indeed, when $H_{0\perp} \gg H_{rf}$ the total field $H = H_{0\perp} + H_{rf}$ oscillates about the direction of $H_{0\perp}$, and $\chi_\perp$ is large. The field $H_{0\parallel} \gg H_{rf}$ restrains the spin rotation induced by H_{rf} ($\chi_\parallel$ is small). Therefore, $H_{0\perp}$ provides the optimum conditions to increase I_{sb}.

In the case of technical magnetization of α-Fe the intensivity I_{sb} is little affected by an increase in $H_{0\perp}$, and drops rapidly in the $H_{0\parallel}$ fields[4]. The dependence of the paraprocess striction on H_0 leads to the fact that for invar the function $I_{sb}(H_{0\perp})$ increases sharply as H_0 increases, and $I_{sb}(H_{0\parallel})$ decreases more slowly than the corresponding function for α-Fe reported in[4].

The spectra obtained for 35% Ni alloy with the continuous-mode pumping at various $H_{0\perp}$ coincide with those taken in the pulsed mode (Fig. 2, a'—d'). The identity of the spectra is evidence that 500 μsec is enough to establish stationary ultrasonic vibrations of the nuclei. No sidebands occur in the absence of the rf field (Fig. 2, a—d), which points to the rapid damping of these vibrations.

A study of the 30% Ni alloy showed small I_{sb}, and the bands were detected only in the presence of $H_{0\perp}$. This is accounted for simply by the fact that for this alloy T_c = 70° C, i.e., substantially lower than T_c for the 35% Ni alloy. Considering that the sample heating was due to the applied rf field, less rf power was required to reach T_c for the 30% Ni alloy (h_{rf} in (2) is small), and, consequently, I_{sb} was small. As the power is further increased, the sample temperature goes above T_c, and the sidebands disappear. To study the rf magnetostriction of such alloys the sample must be cooled in the course of the experiment.

So, the generation of ultrasound in invars by the rf paraprocess striction reaches its maximum near T_c and in the presence of H_0. Under these conditions Hausch and Warlimont[9] have also observed anomalous ultrasound (10 MHz) absorption in invars.

In a recent paper[10] Dubovtsev, Zyryanov, and Filippova reported an observation of the sidebands in Mössbauer spectra of nonferromagnetic metallic samples, including an invar alloy with 36% Ni, at $T > T_c$. The authors ascribe the formation of the bands to eddy currents. However, as stated above, in the invar alloys we have studied, the sidebands disappear when, as the power increases, the samples are heated above T_c. Such a dependence on the field power is inconsistent with an eddy-current model. The discrepancy is likely to be due to a difference in the thickness of the samples. The foils studied in[10] were 4 μm thick, which is comparable to the rf skin depth, while our samples were thicker (25 μm), and this may be responsible for the small value of I_{sb} in our experiments at $T > T_c$. It is conceivable, however, that the sidebands detected in[10] are also due to paraprocess striction. Indeed, as Tino and Maeda h have found, the high-temperature tail of the striction exceeds T_c substantially for an alloy with 35.9% Ni, owing to the short-range magnetic order[11].

Sidebands were also reported[10] in the spectrum of a 3 μm thick sample of paramagnetic stainless steel Kh18N9T. We believe that the ultrasonic vibrations in this case were again due to rf paraprocess striction. Such nonmagnetic steels with γ-phase structure contain a minor amount (≈ 1%) of residual ferromagnetic martensite[12]. They also contain small regions of positive exchange interaction which serve as martensite nucleation sites[13].

For paraprocess striction to be appreciable, a marked dependence of the exchange integral on interatomic separation is required. It has been proposed by Belov[7] that such a dependence occurs in the case when both positive and negative exchange interactions appear in an alloy, in particular near the $\alpha - \gamma$ interphase. This is realized in invar alloys[14, 15], as well as in many stainless steels, including Kh18N9T. In the latter case the ultrasonic vibrations seem first to be generated in the martensite phase and at its nucleation sites, and then spread over the host structure.

The sidebands appear in Mössbauer spectra of Fe^{57} at our value of ν when the ultrasonic displacements of the nuclei become comparable with or exceed the wavelength $\bar{\lambda}$ of the Mössbauer radiation. Knowing that $\bar{\lambda} = 1.4 \times 10^{-9}$ cm for Fe^{57} we can estimate the least value of strain detectable in 35% Ni alloy with $h_{rf} \approx 5$ Oe:

$$\omega=3\frac{\Delta l}{l}\geqslant\frac{3\bar{\lambda}}{d}\sim 2\cdot 10^{-6},\qquad \frac{d\omega}{dH}=\frac{3}{l}\frac{\Delta l}{\Delta H}\sim 4\cdot 10^{-7}$$

(d being the sample thickness). On the other hand, static measurements[11] in 2—7 kOe fields yielded a striction value $\omega = 2 \times 10^{-4}$ (at 6.9 kOe) and $d\omega/dH = 3.5 \times 10^{-8}$ for an alloy with 35.9 at.% Ni.

A comparison of these data gives reason to believe that in weak external fields (≈ 5 Oe) the strain increases faster than in the case of strong fields (≈ 5 kOe). The Mössbauer effect offers a means of observing striction phenomena in weak fields, which makes it possible to avoid the influence of strong magnetic fields on the magnetic state of a sample. This may provide important help to the study of critical phenomena.

Let us also note that comparison of the above estimates with the results of static measurements reveals little frequency dependence of the paraprocess striction in invars.

In conclusion, the authors wish to thank V. I. Gol'danskiĭ and I. M. Puzeĭ for their interest and attention to the work.

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Translated by S. Luryi
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