Polarization of Fe^{57} nuclei when NMR and ferromagnetic-resonance frequencies are equal

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(Submitted 25 April 1981; resubmitted 9 July 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **34**, No. 4, 211–215 (20 August 1981)

A Mössbauer method has been used to detect changes in the resonant frequency of the Fe^{57} nucleus and in the populations of the nuclear sublevels at an intersection of the resonant-frequency branches corresponding to the electronic and nuclear spin systems (in other words, at an intersection of the ferromagnetic-resonance and NMR frequencies) in a thin iron plate in the absence of rf excitation.

PACS numbers: 76.50. + g, 76.60. – k, 75.70.Dp, 76.80. + y

The method of dynamic nuclear polarization in paramagnets involves arranging "direct thermal contact"[1] between the nuclear spin system and an electron spin–spin reservoir, cooled beforehand by microwave pumping. A corresponding effect can apparently be observed in ferromagnets when the resonant frequencies of the nuclear and electronic subsystems are equal. In this case the role of the microwave pump is played by an exchange interaction that polarizes the electron spins. The nuclear spins are also polarized by a resonant energy exchange between the electronic and nuclear subsystems. It is a simple matter to equalize the NMR and ferromagnetic-resonance frequencies in a thin plate[2] with an anisotropy easy axis oriented in the plane of the sample, by applying a static field perpendicular to the anisotropy axis in the same plane.

Figure 1 shows frequency–field curves for this geometry for the homogeneous mode,[2] the low-frequency vibrational branch in the presence of domains,[3] and the lower boundary of the spin-wave frequency.[4] The effective anisotropy field is $H_K = H_A + 4\pi M_0(N_y - N_z)$, where H_A is the anisotropy field, and N_y and N_z are the demagnetizing factors in the plane of the plate. The unperturbed NMR frequency (ω_n) is essentially independent of the static field (the effective field at the Fe^{57} nucleus in iron is 330 kOe). By adjusting the size and shape of the plate (at a constant plate thickness) we can change H_K; as a result, we can also change the number of intersections of the unperturbed vibrational branches of the electronic and nuclear subsystems as well as the positions of these intersections along the H_0/H_K axis.

Changes in the NMR and ferromagnetic-resonance frequencies in an intersection region have been observed previously by rf spectroscopy[5] and by the Mössbauer method[6] with rf pumping. The present experiments made use of the circumstance that the Mössbauer method can be used to measure resonant nuclear frequencies and the populations of nuclear sublevels without the perturbation of the sample which would be caused by applying a resonant rf field.

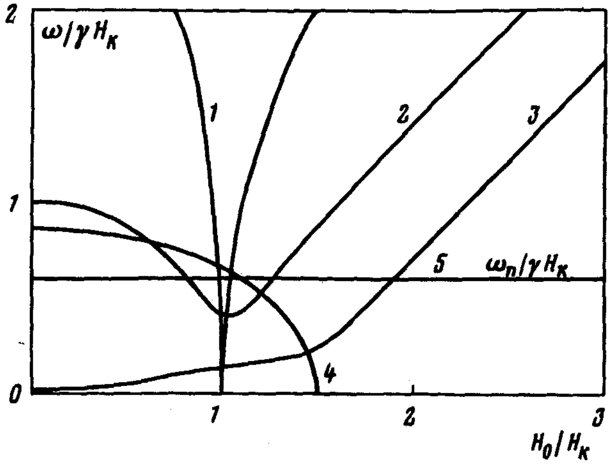

FIG. 1. The resonant frequencies of a ferromagnetic plate vs the magnetic field (H_0). 1–Homogeneous precession (it is assumed that $4\pi M_0/H_K = 10$); 2–lower boundary on the spin-wave frequency for an infinite plate ($N_y = N_z = 0$); 3–lower boundary on the spin-wave frequency at $H_A = 4\pi M_0 N_z = 2\pi M_0 N_y$; 4–"domain" oscillation branch; 5–nuclear frequency.

A constant-acceleration spectrometer[7] was used to observe the Mössbauer spectra of thin plates of pure iron of various shapes (Fig. 2a) with easy axis lying in the plane of the plates (along the z axis). To improve the resolution, we monitored only the inner lines of the Fe^{57} spectrum [corresponding to the transitions $(-1/2)$-$(+1/2)$ and $(+1/2)$-$(-1/2)$]. The interval between the lines corresponds to the difference between the NMR frequencies of the ground and excited states of the nucleus. Since the magnitude of the splitting of the excited state does not change, (even with strong rf pumping,[6] because of the short lifetime and low concentration of excited nuclei), the change in the line spacing upon a change in H_0 is due entirely to a change in the NMR frequency of the ground state. The spectral envelopes (A and B in Fig. 2b) and the parameters of the line were found by numerical calculations using the computer program of Ref. 8.

We determined the relative change in the resonant frequency, $\Delta\omega/\omega_0$ (ω_0 is the NMR frequency with $H_0 = 0$), and the relative population change of the nuclear sublevels, $\Delta S/S_0$ (ΔS is the difference between the areas under the Mössbauer lines, and S_0 is the average area under these lines). A positive value of $\Delta S/S_0$ corresponds to a cooling of the nuclear spin system, while a negative value corresponds to a population inversion, i.e., to a negative spin temperature.

As expected,[2] as H_0 is reduced from the value corresponding to the saturation state of the sample (the region of negative values of H_0 in Fig. 2a) to $H_0 = H_K$, the homogeneous-mode frequency (curve 1 in Fig. 1), which drops to zero near H_K, is equal to the unperturbed frequency of the nucleus ($\omega_n = 2\pi \times 45.44$ mHz); this behavior causes a repulsion of the branches and a change in the resonant NMR frequency. For sample No. 2 (Fig. 2a) we can clearly see two resonant-frequency branches: (quasinuclear and quasielectronic). The distance between the branches is

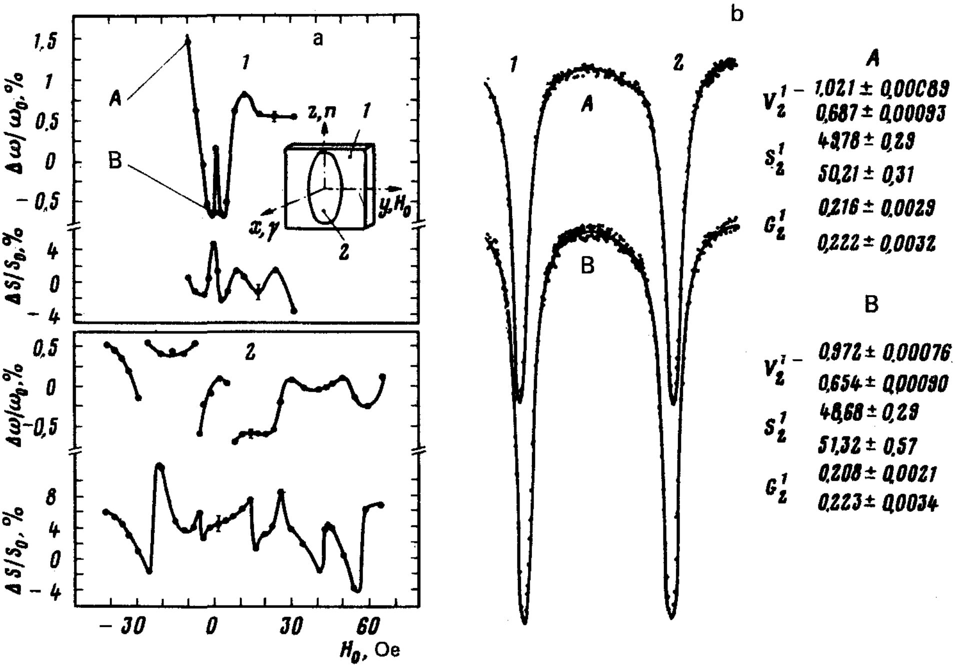

FIG. 2. (a) Relative change in the resonant frequency of the nucleus, $\Delta\omega/\omega_0$, and that in the population of the nuclear sublevels, $\Delta S/S_0$, vs the magnetic field H_0. 1–Square sample [$(5 \times 10^{-3}) \times 15 \times 15$ mm; $N_z \approx N_y$; $H_K = H = 3$ Oe)]; 2–elliptical sample [$(5 \times 10^{-3}) \times 5 \times 10$ mm; $N_y \approx 3N_z$; $H_K = H_A + 8\pi M_0 N_z \approx 25$ Oe]. (b) NMR spectra corresponding to points A and B in Fig. 2a. V–Line position, in millimeters per second, with respect to Co^{57} in a Pd matrix; S–area under the line, as a percentage of the total area; G–line width, in millimeters per second.

determined by the hyperfine interaction parameter. At the same time, at the intersection point, there is an abrupt change in the nuclear polarization (the situation is analogous to the change in the oscillation phase of coupled pendulums). The shape of the curve (the curve of $\Delta S/S_0$ vs H_0 near H_K) is reminiscent of the curve of the reciprocal temperature of the spin–spin reservoir of paramagnetic centers vs the frequency distance from resonance in the case of dynamic cooling.[1] A further decrease in H_0 (from H_K to 0) leads to a homogeneous rotation of the magnetization vector in the plane of the plate, toward the easy axis. A domain structure does not arise[2,9] ($H_K > 4\pi M_0 N_z$). In this field interval, the nuclear branch for sample No. 2 intersects the lower spin-wave boundary (for sample No. 2, $\omega_n/\gamma H_K = 0.63$, and the spin-wave boundary lies slightly below curve 2 in Fig. 1, having the same shape as curve 2). A change in the sign of H_0 leads to a domain structure (at $H_0 \sim 5$ Oe, which is the coercive force) with domain walls running parallel to the easy axis and with the characteristic frequency dependence[3,9] (curve 4 in Fig. 1). The transition from the single-domain state to the multidomain state occurs abruptly, leading to an abrupt change in the nuclear frequency as a result of monotonic change in the polarization. (The branches are repelled but do not intersect. Their intersection point is at ~45 Oe; see

Fig. 2a for sample No. 2.) For sample No. 1 there is no intersection with the domain branch (for this sample, $\omega_n/\gamma H_K = 5.2$), and the intersection with the spin-wave boundary is observed at $H_0 = 24$ Oe. At fields from 5 to 30 Oe the domains distort the spin-wave spectrum,[10] causing the experimental points to deviate from the results of this simple calculation.

Interestingly, the change in the entropy of the nuclear spin system in the region where the NMR and ferromagnetic-resonance frequencies are equal can actually be used to cause cooling (by analogy with the nuclear-demagnetization method).

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Translated by Dave Parsons
Edited by S. J. Amoretty