Investigation of electron–nuclear oscillations in ferromagnetic films under conditions of strong coupling between the electron and nuclear subsystems

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(Submitted 12 September 1978)
Pis'ma Zh. Eksp. Teor. Fiz. 28, No. 10, 621–624 (20 November 1978)

The electron–nuclear branches were observed in experiment to "move apart" when the FMR and NMR frequencies in thin ferromagnetic films coincided. The effect was observed at liquid-helium temperatures, when the interaction between the spin subsystems exceeds the damping of the electron and nuclear magnetizations.

PACS numbers: 76.50. + g, 76.60.Jx, 73.60. – n

We report here an experimental observation, for the first time ever, of repulsion of electron–nuclear oscillation modes in ferromagnetic films as a result of interaction between the electron and nuclear subsystems when the frequency–field dependences of the FMR and NMR intersect.

This effect was predicted by Ignatchenko and Kudenko,[1] and an attempt to observe it experimentally at 300 K was made by us earlier.[2] Instead of the expected "separation" of the electron–nuclear modes, we observed a characteristic "kink" on the FMR frequency–field curve, and an enhancement of the NMR signal in the region where the frequencies became equal. The hypothesis was advanced that allowance for the damping can explain this effect.

A detailed analysis of the conditions for the onset of the "separation," with allowance for the damping in the electron and nuclear subsystems, contained in the theoretical papers of Portis,[3] Botvinko and Ivanov,[4] and Ignatchenko and Tsifrinovich[5] have shown that the necessary condition for the onset of the "separation" of the electron–nuclear modes is the inequality $\omega_x \gg \omega_r$, where $\omega_x = \gamma_e(4\pi AmM)^{1/2}$ is the exchange frequency, ω_r is the rate of relaxation in the electron subsystem, m is the nuclear magnetization, M is the electronic magnetization, and A is the hyperfine interaction constant. If, on the other hand, $\omega_x \approx \omega_r$, "kinks" should be observed on the frequency–field curves, as in Ref. 2.

The inequality $\omega_x \gg \omega_r$ can be realized in ferromagnetic films at low temperatures, since $m \propto 1/T$ K. For Fe–Ni–Co films, the experimentally measured rate of electron relaxation in local regions is approximately 1.3×10^8 sec^{-1}, and the exchange frequency ω_x at 300 K is 2×10^8 sec^{-1} (i.e., $\omega_x \approx \omega_r$ at 300 K), while at 1.5 K it reaches a value 3×10^9 sec^{-1} (i.e., $\omega_x \gg \omega_r$ at 1.5 K). The experiment was performed, as in the preceding cases, on Fe–Ni–Co films of non-magnetostriction composition with about 40% cobalt, but at liquid-helium temperatures. To prevent the antiferromagnetic oxide layer from influencing the measurement results, the films were obtained in ultrahigh vacuum and covered subsequently with a protective coating. The sample was placed in a nonresonant strip line of matched wave resistance. At frequencies below 200 MHz, the measurements were performed by the spin-echo method, and at higher frequencies by absorption of the energy of continuous high-frequency oscillations.

Figure 1 shows the dependence of the echo-signal amplitude on the external magnetic field, obtained with the aid of a integrator and an x-y recorder at 180 MHz, with the exciting pulses separated by 400 μsec and at a temperature 1.5 K. Two maxima are clearly seen on both sides of the magnetic-field value corresponding to the unperturbed FMR. We note that the curve has one maximum already at liquid-nitrogen temperature. A series of similar curves for different frequencies made it possible to plot the electron–nuclear branches with the frequency and magnetic field as coordinates (Fig. 2).

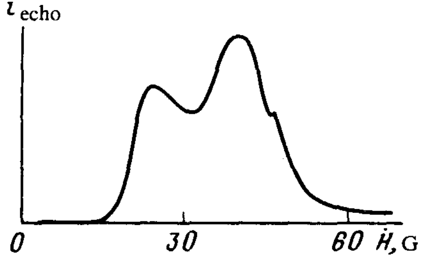

FIG. 1. Dependence of the spin-echo intensity in Fe–Ni–Co films on the external magnetic field. Frequency 180 MHz, temperature 1.5 K, distance between exciting pulses 400 μsec.

In Fig. 2, the solid thin lines show the unperturbed values of the natural frequencies of the system, obtained at room temperature, when the FMR was investigated at high power under conditions of saturation of the nuclear subsystem, and the NMR was investigated by the spin-echo method. The experimental points are given for the temperature 1.5 K, when the "electron-like" section of the branches in the upper part of the figure were measured by a continuous procedure at very low power levels, while the electron–nuclear branches in the lower part of the figure were measured by the spin-echo method. In this case one of the branches of the electron–nuclear oscillations corresponds to interaction of the nuclear subsystem with multidomain FMR, while the other corresponds to interaction with single-domain FMR.

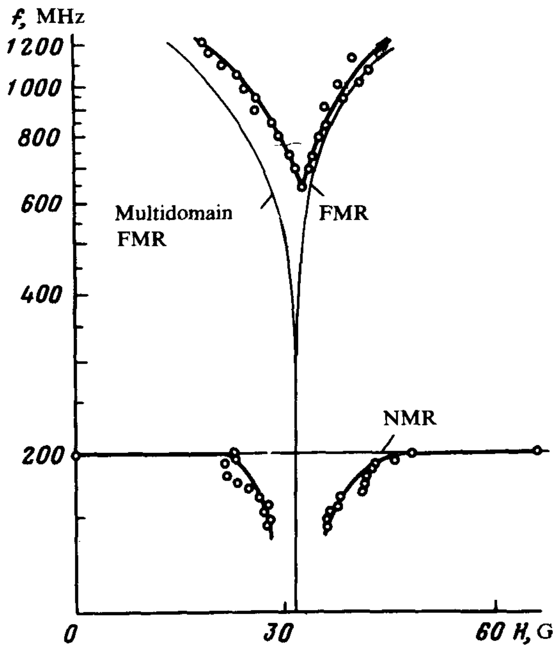

FIG. 2. Dependence of the frequencies of the electron and nuclear magnetic resonances in an Fe–Ni–Co film on the external magnetic field in the absence of coupling between the subsystems (solid thin lines) and in the case of strong coupling at $T = 1.5$ K (experimental points and thick lines).

A characteristic feature of the behavior of the "electron-like" sections of the electron–nuclear branches is the strong dependence of the absorption coefficient on the exciting power and on the temperature: at low exciting-power levels and at low temperatures (below 77 K) the absorption vanishes completely at a certain critical frequency (400–700 MHz, depending on the temperature). On the other hand, at high power levels the ordinary FMR is observed without substantial singularities in the entire interval of the frequencies and temperatures.

The vanishing of the absorption of the high-frequency energy in this case is apparently due to the fact that the upper electron–nuclear branch, which corresponds to the "entanglement" of the multidomain FMR with the NMR, shifts towards magnetic fields stronger than the anisotropy field, i.e., into a region where the multidomain state is destroyed, while the upper branch, which corresponds to the interaction of the single-domain FMR with the NMR, shifts towards fields weaker than the anisotropy field, i.e., into a region where the single-domain state is destroyed.

Thus, the upper branches move towards each other and the point of their intersection determines that limiting frequency below which there is no absorption. Owing to the large slope of the frequency–field dependence of the FMR in the investigated frequency range, the limiting frequency is very sensitive to the temperature and to the power. For this reason, no "nuclear-like" sections of these electron–nuclear branches have been observed in experiment.

The lower electron–nuclear branches are located in regions corresponding to stable multidomain and single-domain states, and therefore lend themselves fully to experimental investigation. The change of the frequency of the echo signals along these branches reached 60 MHz in these experiments. The echo-signal amplitude decreased gradually and measurement at lower frequencies by the pulse procedure became difficult. It should be noted that similar relations can be obtained also at 4.2 K, but with a worse separation of the electron–nuclear branches.

Figure 3 shows the variation of the rate of the transverse relaxation of the electron–nuclear oscillations as a function of the external magnetic field, determined by measuring the falloff of the echo signals with increasing distance between the exciting pulses. The data are presented for 1.5 K and 180 MHz. As seen from Fig. 3, the behavior of the rate of transverse relaxation is similar to that in Fig. 1. The maxima of the damping practically coincide with the values of the natural frequencies of the coupled electron–nuclear oscillations.

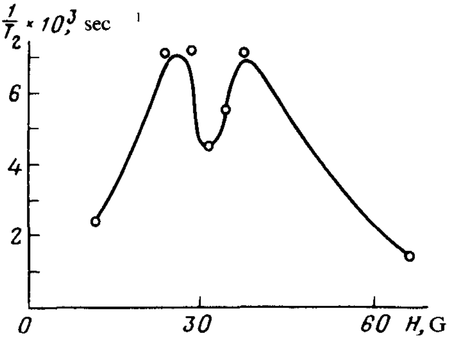

FIG. 3. Rate of transverse relaxation (T_2^{-1}) of the "nuclear-like" oscillations in an Fe–Ni–Co film against the external magnetic field at $T = 1.5$ K and 180 MHz.

Finally, we compare now the obtained experimental data with some theoretical conclusions. According to the data of Portis, as well as that of Ignatchenko and Tsifrinovich, the maximum value of the separation is ω_x, meaning that at 1.5 K we have $\omega_x/2\pi \approx 480$ MHz. According to Botvinko and Ivanov the separation is given by the formula

$$\omega_x = \sqrt{cn}\,\omega_0 ,$$

where c is the concentration of the magnetic nuclei, n is the parameter of dynamic coupling, approximately $14/T$ K for cobalt nuclei, and ω_0 is the frequency of the unperturbed NMR. For $T = 1.5$ K and $c = 0.4$ this amounts to approximately 400 MHz. The separation of the electron–nuclear branches in our experiment can be calculated from an analysis of the curves in the lower part of Fig. 2. If it is assumed that one end of each electron–nuclear branch shown there approaches the unperturbed FMR at zero frequencies, and the other end approaches the frequency of the unperturbed NMR, then it is seen that the separation does not exceed the maximum theoretically calculated value.

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