Effect of resonant RF field on the hyperfine structure of the nuclear levels in a paramagnetic crystal

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(Submitted May 16, 1977)
Pis'ma Zh. Eksp. Teor. Fiz. **26**, No. 1, 16–19 (5 July 1977)

Results are reported of an investigation of the Mössbauer effect on Fe^{57} nuclei in the paramagnet $Al(NO_3)_3 \cdot 9H_2OFe^{3+}$ following simultaneous action of a constant and alternating magnetic field. It is shown that a radiofrequency (RF) field of resonant frequency causes an appreciable broadening of the Mössbauer spectrum components, due to the splitting of the sublevels of the excited state of the Fe^{57} nucleus.

PACS numbers: 76.80.+y, 71.70.Jp

In 1961, Hack and Hamermesh[1] calculated the change in the shape of the Mössbauer line in an alternating field whose frequency is equal to the frequency of the hyperfine structure of the excited state of the nucleus. Their main result was that in sufficiently strong RF fields each spectral component splits into $2I_e+1$ components, where I_e is the spin of the excited state of the nucleus. A similar result was obtained later by Gabriel.[2]

It was shown in[1,2] that the broadening and splitting of the Mössbauer-spectrum components becomes noticeable only when the RF field amplitude is comparable with the width of the Mössbauer line ($H_{RF} \approx \Gamma$). For Fe^{57} nuclei, e.g., H_{RF} should be no less than 10 kOe at frequencies on the order of several dozen MHz. Obviously, such fields cannot be obtained at present under laboratory conditions.

It is known, however, that in ferromagnetic materials there exists a mechanism of enhancing the RF field via exchange interaction, as a result of which the intensity of the effective RF field acting on the nucleus is sufficient to satisfy the condition $H_{eff} \approx \Gamma$.

However, attempts to observe this phenomena in experiments (in a geometry where the resonant Mössbauer γ photons are absorbed), in the form predicted in[1,2], did not yield positive results.[3] The point is that the onset of magnetostriction oscillations in the magnetically ordered samples following applica-

 0021-3640/77/2601-0013$00.60

tion of H_{RF} leads to the appearance of a large number of "satellites" in the Mössbauer spectrum.[4] This hinders appreciably its interpretation and does not make it possible to draw an unambiguous conclusion concerning the influence of the RF field directly on the structure of the nuclear levels.

Moreover, special investigations undertaken with an aim at observing the effect in experiments on resonant scattering of γ photons in magnetically ordered substances have shown that even in those case when such an influence does take place, the observed magnitude of the effect turns out to be much less than that predicted by the theory.

In paramagnetic crystals, under certain conditions, it is also possible to enhance the RF field via a hyperfine electron-nuclear interaction.[6] It can be shown that the gain in such crystals for the Fe^{57} nuclei reaches $\sim 10^3$. The absence of magnetostriction oscillations in paramagnets, at a gain of the same order of magnitude as in magnetically-ordered substances, makes them preferable objects for the investigation of the new physical phenomenon discussed here.

We have investigated the effect of an RF field on resonant absorption of γ rays of energy 14.4 keV by Fe^{57} nuclei, using a Mössbauer spectrometer operating in the constant source velocity regime (Co^{57} in Cr, activity 100 mCi). The absorber was an aluminum nitrate $Al(NO_3)_3 \times 9H_2O:Fe^{3+}$ single crystal, with iron-impurity ion (90% Fe^{57}) concentration not larger than 0.5 mol.%.[7,8] The RF field H_{RF} was produced by the coil of a special independently excited oscillator whose output stage was a tuned power amplifier.[9] The investigated sample, in a stream of liquid nitrogen, was placed in an organic-glass vessel connected to a metallic cryostat. The oscillator coil, with inductance $L \approx 1\ \mu$H, was placed in the vacuum volume of the cryostat and was mounted on the sample holder in such a way that the sample itself was at the center of the coil, whose axis coincided with the direction of the γ-ray beam. The constant magnetic field H_0 applied to the sample perpendicular to the γ-ray beam was produced by two ferrite rings secured on the outside of the cryostat.

It was established earlier[7,8] that the Mössbauer absorption spectra of the Fe^{57} nuclei in this crystal, when placed in a stabilizing magnetic field $H_0 \geq 100$ Oe at $T \leq 77$ K, are superpositions of Zeeman spectra consisting of three Kramers doublets into which the electron spin of the Fe^{3+} ion is split in the crystal field. For one of these doublets, the hyperfine interaction tensor A_{jk} is fully isotropic, so that the field $H_{hf} = 255$ kOe for the Zeeman spectrum corresponding to this doublet (lines 1 to 6—see Fig. 1) is always parallel to $\mathbf{H}_0$. It can be shown that in this case the effective alternating field acting on the Fe^{57} nucleus is $H_{eff} \approx H_{RF}(H_{hf}/H_0)$. At $H_0 = 100$ Oe and $H_{RF} = 10$ Oe the field intensity $H_{eff} = 25$ kOe is commensurate with the experimental width of the Mössbauer spectrum components in $Al(NO_3)_3 \times 9H_2O:Fe^{3+}$ ($\Gamma_{exp} \approx 20$ kOe). Under these conditions, the action of an RF field of resonant frequency ($\nu_{res} = 19$ MHz for the excited state of the nucleus in the Zeeman spectrum of interest to us) will lead only to a broadening of lines 1—6. The lines corresponding to two coinciding "anisotropic" Kramers doublets should not undergo any noticeable change in this case.[8]

It is seen from Fig. 1 that application of the RF field changed precisely this part of the spectrum. The lines 1—6 were broadened by more than 1.5 times,

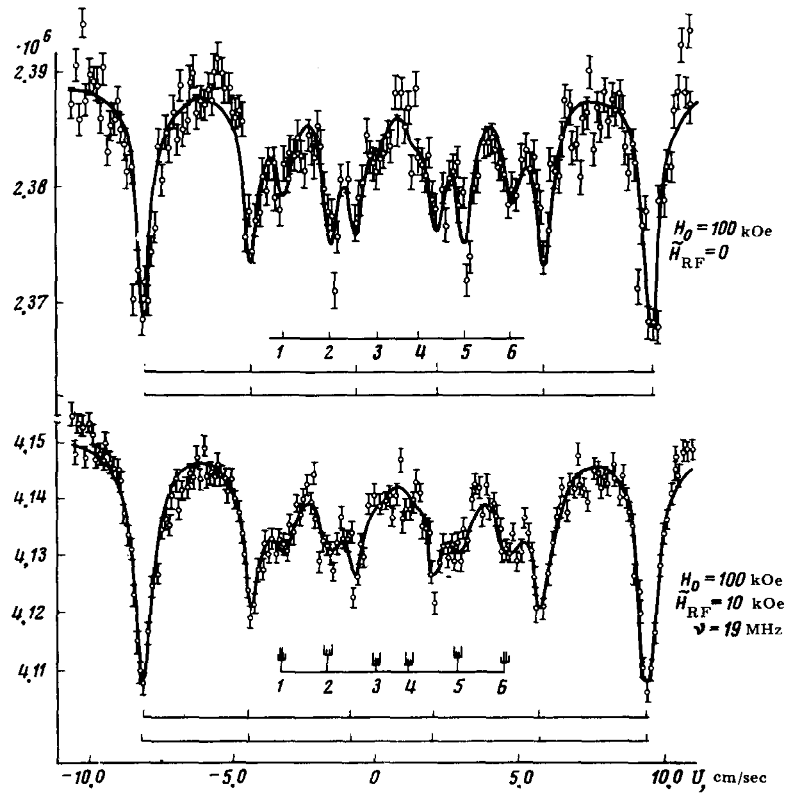

FIG. 1. Mössbauer spectra of the Fe^{57} nuclei in $Al(NO_3)_3 \times 9H_2O: Fe^{3+}$ at $T = 77$ K in a constant (H_0) and radiofrequency (H_{RF}) magnetic fields ($\mathbf{H}_{RF} \perp \mathbf{H}_0$). The source is Co^{57} in Cr ($T = 300$ K).

whereas the remaining lines remained practically unchanged. It is important that the influence of the RF field manifested itself in most pronounced form for the components 2 and 5, since the character of the splitting (the intensity ratio) of these lines leads to a larger broadening (compared with components 1 and 6) of their envelope in the resultant spectrum. The influence of the RF field has thus in this case a "selective," resonant character, and the magnitude of the observed effect agrees qualitatively with the conclusions of the theory. [1,2] On the other hand, to observe visually the splitting of individual spectral components it is obviously necessary to increase greatly the γ-ray statistics.

The analysis of the results indicates that in the crystal $Al(NO_3)_3 \times 9H_2O: Fe^{3+}$ it is relatively easy to realize the conditions necessary for the experimental observation of the resonant action of the RF field on the hyperfine structure of the nuclear levels of Fe^{57}. This fact offers considerable promise for investigations aimed at a detailed study of the resonant phenomena in paramagnets.

The authors are deeply grateful to I. K. Kikoin for constant interest and

attention and to A. M. Afanas'ev for very useful discussions of the formulation of the problem and of the experimental results.

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