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#### INFLUENCE OF CROSS RELAXATION PROCESSES ON THE $\gamma$ RESONANCE SPECTRA OF $\text{Fe}^{57}$

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We report below a study of the influence of the cross relaxation processes on the  $\gamma$  resonance spectra of  $\text{Fe}^{57}$  in two model systems, constituting solutions of complexes produced in n-butyl alcohol in which ferric chloride is dissolved. In the case of complexes of the first type ( $K_1$ ) the splitting of the spin sublevels of the  $\text{Fe}^{3+}$  ions by the crystal field does not exceed  $10^{-2} \text{ cm}^{-1}$ , whereas in the complexes  $K_2$  the splitting is much larger and equals  $2 \times 10^{-1} \text{ cm}^{-1}$ . The distances between the spin sublevels were estimated from the EPR spectra. The concentrations of the complexes  $K_1$  and  $K_2$  (Fig. 1a, 1b, 1c, and 2a, 2b;  $T = 88^\circ\text{K}$ ) were  $5 \times 10^{19} \text{ cm}^{-3}$  and  $10^{20} \text{ cm}^{-3}$ , respectively. The rapidly-relaxing magnetic ion introduced in the solutions of the complexes was  $\text{Co}^{2+}$ .

It is known that cross relaxation processes that include a simultaneous change of the projection of the electron spin of two or more neighboring ions take place under certain conditions in a system containing two or more types of paramagnetic ions. Such processes have the greatest probability if the energy is strictly conserved in the spin system, i.e., the conditions  $n\Delta E_{ab} = m\Delta E_{a'b'}$ , are satisfied, where  $n$  is the number of particles going from level  $a$  to level  $b$ ,  $m$  the number of particles going from level  $b'$  to level  $a'$ , and  $\Delta E_{ab}$  and  $\Delta E_{b'a'}$ , the energy intervals between the corresponding levels.

The greatest probability is possessed by the process of two-spin (resonant) cross relaxation ( $n = m = 1$ ). The probability of such processes is determined by the proximity of the energy intervals between the spin sublevels of the interacting pair of paramagnetic ions. One can therefore expect the  $\text{Co}^{2+}$  ions to act differently on the complexes  $K_1$  and  $K_2$ , which have different Stark splittings. To improve the resolution of the hyperfine structure of the  $\gamma$ -resonance spectra, the samples were placed in external magnetic fields [2].

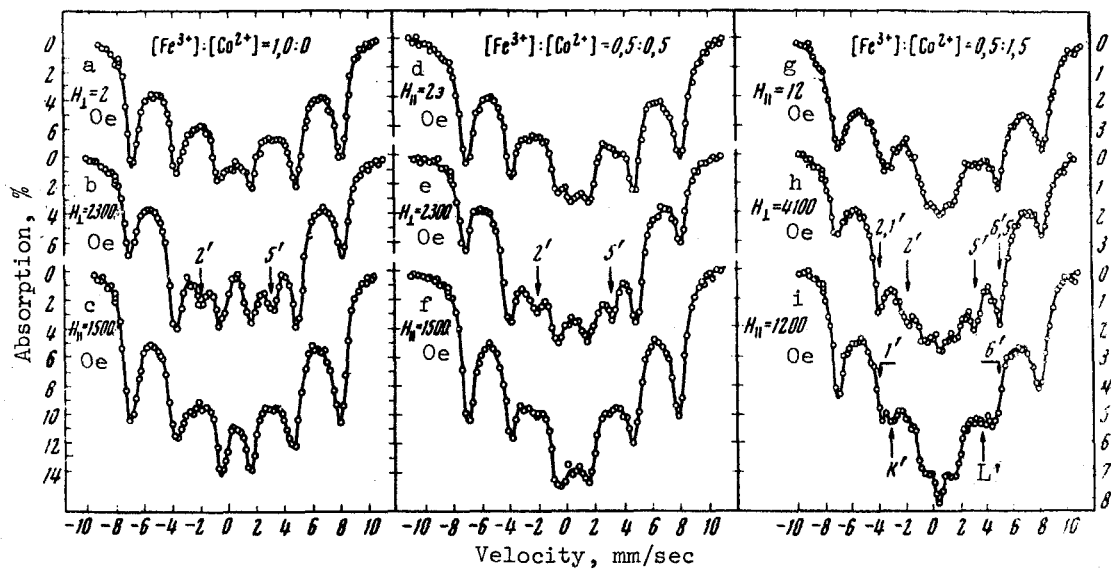


Fig. 1. Gamma-resonance spectra of frozen solutions of the complex  $K_1$  in n-butyl alcohol at different external magnetic fields and at different concentrations of the ions  $Fe^{3+}$  and  $Co^{2+}$ : a, b, c -  $[Fe^{3+}] = 5 \times 10^{19} \text{ cm}^{-3}$ , d, e, f -  $[Fe^{3+}] = [Co^{2+}] = 2.5 \times 10^{19} \text{ cm}^{-3}$ ; g, h, i -  $[Fe^{3+}] = 2.5 \times 10^{19} \text{ cm}^{-3}$ ,  $[Co^{2+}] = 7.5 \times 10^{19} \text{ cm}^{-3}$ ;  $T = 88^\circ\text{K}$ .

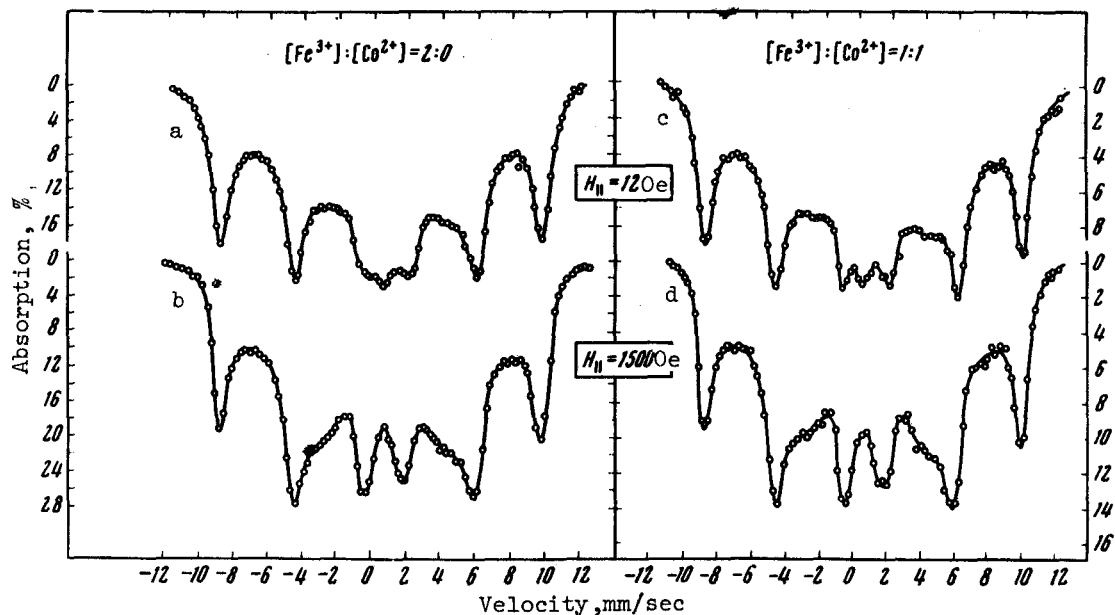


Fig. 2. Gamma-resonance spectra of frozen solutions of the complex  $K_2$  in n-butyl alcohol at different values of the external magnetic field, with and without  $Co^{2+}$  ions: a, b -  $[Fe^{3+}] = 10^{20} \text{ cm}^{-3}$ ; c, d -  $[Fe^{3+}] = [Co^{2+}] = 5 \times 10^{19} \text{ cm}^{-3}$ .  $T = 88^\circ\text{K}$ .

It is obvious from the spectra shown in Fig. 1 that introduction of the  $\text{Co}^{2+}$  ions in the solution of the complex  $\text{K}_1$  influences the relaxation processes of the iron ions. In the central parts of the spectra (Figs. 1d, 1e, and 1f [ $[\text{Fe}^{3+}] = [\text{Co}^{2+}] = 2.5 \times 10^{19} \text{ cm}^{-3}$ ]) there appears additional absorption, evidently indicating acceleration of the  $\text{Fe}^{3+}$  ion relaxation. We note that in this case the concentration of the Fe ions is decreased to one-half the concentration of the sample whose spectra are shown in Figs. 1a, 1b, and 1c. Consequently, the observed acceleration of  $\text{Fe}^{3+}$ -ion spin relaxation can be attributed only to the action of the  $\text{Co}^{2+}$  ions on the relaxation of the  $\text{Fe}^{3+}$  ions.

Figures 1g, 1h, and 1i show the spectra of the solution of the complex  $\text{K}_1$ , in which the concentration of the Co ions was increased threefold, and the Fe-ion concentrations were the same as in Figs. 1d, 1e, 1f: [ $\text{Co}^{2+}$ ] =  $7.5 \times 10^{19} \text{ cm}^{-3}$ , [ $\text{Fe}^{3+}$ ] =  $2.5 \times 10^{19} \text{ cm}^{-3}$ . At such a Co-ion concentration one observes an even more noticeable acceleration of the relaxation of the  $\text{Fe}^{3+}$  spins, as is manifest by a much larger broadening of the components of the magnetic hyperfine structure of the spectra. It is obvious here that both the components of the doublet  $\pm 3/2$  (for example 2' and 5', Figs. 1b, e, h) and of the doublet  $\pm 5/2$  (1 and 6, Figs. 1c, f, i) become broadened.

The increased probability of the cross relaxation processes with increasing concentration of one of the paramagnetic ions [ $\text{Co}^{2+}$ ] is due to the broadening of the spin levels of the ions as a result of the dipole-dipole interaction between them and the increased "overlap" of the resonances of the iron and cobalt ions [3].

Although it is quite difficult to determine the absolute value of the relaxation time from the  $\gamma$ -resonance spectra, nevertheless it can be concluded from a comparison of the spectra of Figs. 1e and 1h that when the  $\text{Co}^{2+}$  concentration is increased threefold the rate of the relaxation times does not increase by more than 2 - 3 times. The transitions predominant in this case are obviously the resonant two-spin transitions, whose intensity depends linearly on the concentration of the paramagnetic impurity [1].

The  $\gamma$ -resonance spectra of a solution of the other complex ( $\text{K}_2$ ), into which  $\text{Co}^{2+}$  ions have been introduced ([ $\text{Co}^{2+}$ ] = [ $\text{Fe}^{3+}$ ] =  $5 \times 10^{19} \text{ cm}^{-3}$ ), are shown in Figs. 2c and 2d. It is seen that these spectra do not differ from the spectra (Figs. 2a and 2b) of a solution containing no Co. We note that the concentrations of the paramagnetic  $\text{Fe}^{3+}$  and  $\text{Co}^{2+}$  ions are in this case double those in which the influence of the cross relaxation on the  $\gamma$ -resonance spectra is already noticeable in the  $\text{K}_1$  complexes (Figs. 1d, e, f). It is obvious that neither in a "zero" magnetic field nor at  $H_{\text{ext}} = 1500 \text{ Oe}$  do the  $\text{Fe}^{3+}$  ions in the  $\text{K}_2$  complexes have levels that are equidistant with the  $\text{Co}^{2+}$  ion, and there is no cross relaxation.

It was reported in [4] that the authors of that reference were unable to observe with the aid of nuclear  $\gamma$ -resonance spectroscopy the cross relaxation processes in a system containing  $\text{Fe}^{3+}$  and  $\text{Co}^{2+}$  ions. In our opinion the absence of cross-relaxation processes in this system is due to the fact that the  $\text{Fe}^{3+}$  and  $\text{Co}^{2+}$  ions in it have no close resonant transitions, as in the particular case of the  $\text{K}_2$  complexes considered above.

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