

## Observation of exchange-induced orthogonal state of magnetic impurity

A. M. Balbashov, A. G. Berezin, Yu. V. Bobryshev, P. Yu. Marchukov,  
I. V. Nikolaev, and E. G. Rudashevskii

*Institute of General Physics, Academy of Sciences of the USSR*

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An impurity mode whose frequency has an unusual dependence on the magnetic field has been observed experimentally in a study of antiferromagnetic resonance in  $\text{YFeO}_3$  at 12 K in magnetic fields up to 120 kOe in the wavelength interval 4–0.75 mm. This unusual magnetic field dependence is a consequence of the orthogonal state of the impurity, whose magnetic moment is oriented strictly antiparallel to the ferromagnetism vector.

Magnetic impurities in antiferromagnets have been studied widely and in detail (see, for example, the reviews<sup>1,2</sup>). In a study of antiferromagnetic resonance (AFMR) in an antiferromagnet with a slight ferromagnetism, however, we have observed an unusual dependence of the frequency of an impurity mode on the magnetic field. The experiments were carried out on a single crystal of yttrium orthoferrite,  $\text{YFeO}_3$ , grown by zone melting with radiative heating<sup>3</sup> from a starting material produced by sintering yttrium oxide,  $\text{Y}_2\text{O}_3$ , and ferric oxide,  $\text{Fe}_2\text{O}_3$ , at 1400 °C. The oxides were mixed in proportions of 1.00:1.00 (per formula unit) within an error of  $10^{-3}\%$ . The total mass fraction of magnetic impurities in the starting components did not exceed  $10^{-4}$  for the  $\text{Fe}_2\text{O}_3$  and  $10^{-6}$  for the  $\text{Y}_2\text{O}_3$ , according to the technical specifications. In addition, we carried out an x-ray fluorescence analysis of the single crystal, after it was grown, in order to detect any magnetic impurities present. That analysis revealed that there were

no impurities of rare earths, cobalt, or copper within an error of  $10^{-5}$  and that the mass fraction of manganese did not exceed  $2 \times 10^{-5}$ , in agreement with the technical specifications. To reduce the number of defects and to keep the  $\text{Fe}^{2+}$  and  $\text{Fe}^{4+}$  ions in the sample, we annealed the sample at  $1400^\circ\text{C}$  in oxygen (20 atm) for 8 h.

We used this sample to study the magnetic resonance spectra at a temperature of 12 K in magnetic fields up to 120 kOe. The field was directed along the  $c$  axis of the orthorhombic crystal. We measured the relative intensity of the radiation of a given frequency (in the interval 75–400 GHz) which was transmitted through the sample as a function of the magnetic field.

In our experimental geometry (the ferromagnetism vector  $\mathbf{M}$  was parallel to, and the antiferromagnetism vector  $\mathbf{L}$  perpendicular to, the magnetic field; see the inset in Fig. 1), no spin reorientation was induced by the magnetic field.

The experimental results are also shown in Fig. 1. In addition to the AFMR which is ordinarily observed,<sup>5</sup> and which corresponds to a quasiferromagnetic mode (the ascending branch), we observe an additional absorption line (the descending branch) in the magnetic field. The intensity of this line is lower than that of the AFMR lines by a factor of two or three, and its width is larger by an equal factor. We did not observe this additional line at liquid-nitrogen temperature.

Although we were studying a high-quality single crystal (with an impurity content as low as  $10^{-5}$ ), we believe that the descending branch which we are observing is

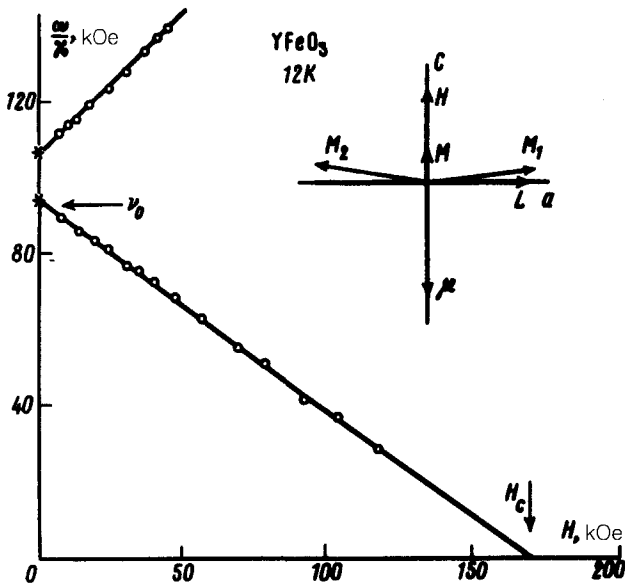


FIG. 1. Resonant frequencies versus the magnetic field. The inset shows the relative orientations of the magnetic field, the sublattice magnetizations ( $M_1$  and  $M_2$ ), and the impurity magnetization  $\mu$ . The asterisks (\*) are data from Ref. 7.

an impurity mode and that the fairly respectable intensity of the lines can apparently be attributed to the "exchange intensification" observed in Ref. 6.

The behavior of the impurity mode as a function of the magnetic field can be described very accurately by the linear function

$$\nu = \nu_0(1 - H/H_c)$$

where  $\nu_0 = 260 \pm 5$  GHz and  $H_c = 170 \pm 3$  kOe.

The value of  $\nu_0$  corresponds to that reported in Ref. 7, where an unknown absorption line was observed in a study of AFMR in YFeO<sub>3</sub> at low temperatures in the absence of a magnetic field. That line was attributed to magnetic impurities in Ref. 7 also.

We wish to stress that in our experiments we observe only the descending branch; there is no additional ascending branch in this frequency range. It can be concluded from the latter result that the magnetic moments of the impurity are directed antiparallel to the magnetic field and thus orthogonal with respect to the antiferromagnetism axis (see the inset in Fig. 1).

What positions might impurity ions be occupying in the crystallographic cell if their magnetic moments are oriented opposite the external field? Let us assume that since the impurity concentration is low, the magnetic moments of the impurity are not coupled with each other by the exchange interaction and that the strength and symmetry of the field acting on an impurity are determined entirely by the magnetic Fe<sup>3+</sup> ions. A situation in which the magnetic moments of an impurity are coupled only weakly with the Fe<sup>3+</sup> magnetic moments, and their orientation is determined exclusively by the anisotropy, evidently could not occur, since in this case one should observe either a single ascending branch (a slight anisotropy) or two branches, one descending and one ascending, which coincide at  $H = 0$ . However, that is not what is seen experimentally. Impurity ions might take one of three positions: They might replace iron ions in 4Fe positions; they might take interstitial positions; or they might replace yttrium ions in 4R rare-earth positions. The magnetic moment of the impurity in the 4Fe positions should be oriented parallel or antiparallel to one of the Fe sublattices (a "collinear" impurity<sup>8</sup>). In the case of an antiparallel arrangement, the magnetic moment of the impurity would indeed have a component directed opposite the slight ferromagnetic moment. When a magnetic field is applied (in all cases, we have  $\mathbf{H} \parallel \mathbf{c}$ ; Fig. 1), this component may decrease, but the decrease in the frequency of the impurity mode will be negligible since the external field cancels out with only the  $\mathbf{c}$  projection of the exchange field. The second possibility—that impurity ions would take interstitial positions—seems unlikely since such an impurity would have to assume regular and furthermore rather symmetric positions, in which the effective field is directed along the  $\mathbf{c}$  axis, if comparatively narrow and intense lines were to be generated.

The situation might be different if the impurity ion occupied 4R positions. In these positions, the symmetry would allow an orientation of the exchange field only along the direction of the  $\mathbf{c}$  axis,<sup>9</sup> and the magnetic moment of the impurity might be directed either along or opposite the slight ferromagnetic moment of the Fe subsystem. Only in the latter case should we observe a decrease in the frequency of the impurity

mode with increasing field. We note in particular that the role of impurity ions might also be played by  $\text{Fe}^{3+}$  ions which have occupied 4R positions. An important point is that the concentration of such ions might be incomparably higher than  $10^{-5}$  but still beyond detection by chemical or x-ray analysis. As a result, in an essentially "impurity-free" crystal (with a magnetic-impurity concentration as low as  $10^{-5}$ ) incomparably an impurity mode would arise.

In our opinion, the probable reason for the observed features of the impurity mode in  $\text{YFeO}_3$  is thus the presence of  $\text{Fe}^{3+}$  ions which have occupied 4R positions in the crystal and whose magnetic moment is directed opposite the slight ferromagnetic moment (perpendicular to the antiferromagnetism axis). We repeat the orthogonal state of the impurity is caused by the direction of the exchange field at the 4R position and is therefore of exchange origin. For this reason, the situation is fundamentally different from that in  $\text{CoCO}_3$  with an Fe impurity,<sup>10</sup> in which the orthogonality of the impurity is a consequence of the (easy-axis) anisotropy of the  $\text{Fe}^{3+}$  ion in the  $\text{CoCO}_3$  matrix (an easy-plane antiferromagnet).<sup>11</sup>

The impurity ions which are ordinarily studied are in either isomorphic substitution positions or interstitial positions. In our case, in contrast, the impurity occupies regular 4R positions. Generally speaking, this circumstance makes it possible to work from AFMR experiments to find the strength of the effective field at the positions of the  $\text{R}^{3+}$  ions.

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