

# Coherent rearrangement of the spin-wave spectrum of an antiferromagnetic cobalt fluoride with a manganese impurity ( $\text{CoF}_2 + 4 \times 10^{-3} \text{ Mn}^{2+}$ )

V. M. Naumenko, V. V. Eremenko, V. M. Bandura, and V. V. Pishko  
*Physicotechnical Institute of Low Temperatures, Ukrainian Academy of Sciences*

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It was experimentally demonstrated for the first time that the impurity concentration is responsible for the rearrangement of the spin-wave spectrum in antiferromagnetic crystals with impurities: the rearrangement of the spectrum is coherent in  $\text{CoF}_2$  crystals with an  $\text{MnF}_2$  impurity of about  $4 \times 10^{-3}$ . The interaction constants of the impurity levels with AFMR  $m_1 \approx 20 \text{ cm}^{-1}$  and  $m_2 \approx 21.5 \text{ cm}^{-1}$  were determined.

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The experimental data for impurity magnetic excitations in crystals with a small impurity concentration ( $c \ll 1$ ), which were obtained earlier, have been satisfactorily explained by the theory, if we can assume that they are localized (see, for example, review article<sup>1</sup>). However, the results of two experimental studies<sup>2,3</sup> have not been explained by the current theory. A line splitting of an antiferromagnetic resonance (AFMR) was observed in one of these investigations<sup>2</sup> and a significant increase of the impurity oscillations as a result of their convergence with the AFMR line was observed in the other investigation.<sup>3</sup> This picture strongly resembled the one predicted by Rashba<sup>4</sup> for an impurity absorption near the excitonic bands in molecular crystals.

A theory,<sup>5-7</sup> which explains this experiment, has recently been developed. This theory is based on the properties of systems with impurity states of large radius. An important property of such systems is their small parameter  $c_0 \approx 3/4\pi \cdot v/r^3$ , where  $v$  is the volume of the unit cell and  $r$  is the radius of the impurity state;  $r$  increases as the impurity level approaches the spin-wave band of the crystal:  $r = a\sqrt{|\Omega/\omega_{\text{imp}} - \omega_{\text{AFMR}}|}$ , where  $a$  is the lattice constant,  $\Omega$  is a value approximately equal to the width of the spin-wave band, and  $\omega_{\text{imp}}$  and  $\omega_{\text{AFMR}}$  are the frequencies of the impurity oscillation and of the AFMR. A collective rearrangement of the spin-wave spectrum begins when  $r \gtrsim r_{\text{imp}}$ , where  $r_{\text{imp}}$  is the average distance between the impurity ions. This rearrangement can be coherent (CR) or incoherent (IR). In the case

of CR the spectrum will have two, mutually separated regions of band states, which are characterized by the wave vectors. The impurity oscillations, no longer localized, become impurity spin waves. In the case of IR a broad region, which is smeared out near the band's edge, is filled by fluctuational localized states; in this case there is only one region of band states. In the case of CR and IR the intensity of the impurity oscillations increases due to natural oscillations, if the impurity oscillations approach the bottom of the spin-wave band.

The most important and rather unexpected result of the new theory is that the rearrangement of the spectrum (CR or IR) is determined by the impurity concentration, although, as usual,  $c \ll 1$ . There is a critical concentration  $c_{crit} = \eta(m/\Omega)^6$ , where  $m$  is the resonance interaction constant of the impurity and matrix oscillations and  $\eta$  is a numerical factor equal to  $\approx 0.1$ , such that if  $c_0 < c < c_{crit}$ , then IR will occur and if  $c > c_{crit}$ , then CR will occur. Our task is to verify this assertion.

A convenient object for investigation is a  $\text{CoF}_2 + \text{Mn}^{2+}$  crystal in which CR and IR were observed earlier.<sup>3,6,8</sup> Since the CR and IR occurred at different orientations of the external magnetic field and different polarizations, an unambiguous conclusion about the reason for the different rearrangement of the spectrum could not be made. We were primarily interested in the region of intersection of the upper Zeeman impuri-

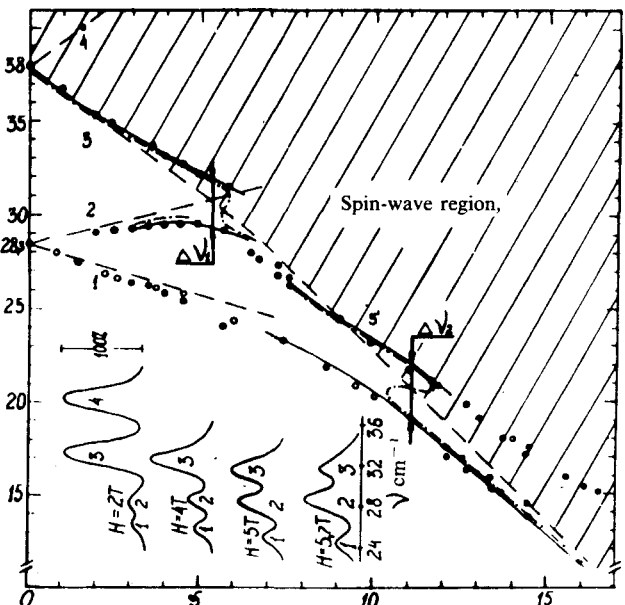


FIG. 1. Behavior of the absorption lines in  $\text{CoF}_2 + 4 \times 10^{-3} \text{Mn}^{2+}$ .  $H \parallel C_4$ —1 and 2, impurity resonance; 3, 3', and 4, AFMR. The dashed curve represents the phenomenological calculation disregarding the interaction between the oscillations; the solid curves and the dot-dash curves represent the theoretical calculations taking into account the oscillations in the cross section with respect to the frequency and the field, respectively;  $\bullet$ , experimental points obtained by frequency scanning of the spectrum;  $\circ$ , the same points obtained by field scanning.

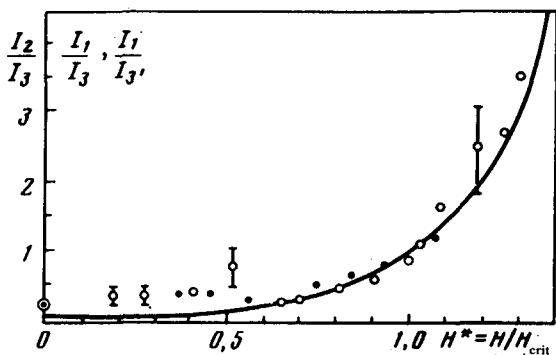


FIG. 2. Dependence of the ratio of the integral line intensities on  $H$ .  $\odot$ ,  $I_2/I_3$ ;  $\circ$ ,  $I_1/I_3$ ,  $I_1/I_2$ ;  $\bullet$ ,  $I_1$  and  $I_2$ —the intensities of the impurity modes 1, 2 in Fig. 1;  $I_3$  and  $I_3'$ —the same for mode 3 of AFMR. The solid curve represents the theory.<sup>7</sup>

ty level with the low-frequency (LF) mode of the AFMR, where an IR in a crystal with a concentration of the  $\text{Mn}^{2+}$  impurity  $c = (2 \pm 1) \times 10^{-4}$  (by weight) was observed earlier.<sup>3</sup> We have investigated a crystal in which  $c = (4 \pm 2) \times 10^{-3}$ . Figure 1 shows a frequency-field dependence of the impurity modes and of the AFMR obtained by us. The evolution of the absorption lines as a result of frequency scanning of the spectrum is shown schematically in the lower left-hand corner. Figure 2 demonstrates the variation of the integral line intensities. We can see from these figures that singularities characteristic of CR can be observed in both regions of intersection of the impurity lines with the LF mode of the AFMR: a cross-splitting of the impurity lines and of the AFMR lines is accompanied by a transfer of the intensity; the interacting lines are divided by an energy interval  $\Delta\nu$  at the point of equality of the intensities ("crossover point"); both lines are clearly visible before and after the crossover point. The coordinates of the crossover points are:  $\nu_{\text{crit}_1} = 30.5 \pm 0.3 \text{ cm}^{-1}$ ,  $H_{\text{crit}_1} = 5.3 \pm 0.2T$ ,  $\nu_{\text{crit}_2} = 20.5 \pm 0.5 \text{ cm}^{-1}$ , and  $H_{\text{crit}_2} = 11.1 \pm 0.5T$ . The lines merged in the case of IR.<sup>3</sup>

The experiment was performed using a long-wave IR diffraction spectrometer and a submillimeter feed-through spectrometer. The samples of good optical quality had the shape of a disk  $\phi = 3\text{--}6 \text{ mm}$  and a thickness of  $0.1\text{--}0.2 \text{ mm}$ . At  $H = 0$  the width of the impurity line and of the AFMR line was about  $1.5 \text{ cm}^{-1}$  and  $2.2 \text{ cm}^{-1}$ , respectively, and did not vary substantially with varying field. The shape of the lines is somewhere between Lorentzian and Gaussian. The concentration of the impurities was determined by a spectral analysis with a calibration of the apparatus according to the sample powders prepared from pure  $\text{CoF}_2$  and  $\text{MnF}_2$  by a weight method.<sup>1</sup> In calculating the intensities, we introduced corrections for the instrumental function of the spectrometers and for the line truncation, so that the error in determining the integral intensity of the lines did not exceed 30%.

The theoretical dependences in Fig. 1 were obtained by the authors<sup>7</sup> with use of the value  $c = 3.5 \times 10^{-3}$ , which gives the best description of the given experiment. Using the expression  $\Delta\nu = 2m\sqrt{c}$ , which is valid for  $c \gg c_{\text{crit}}$ ,<sup>5-7</sup> we determine the reso-

nance interaction constants for the upper and lower crossover [ $\Delta\nu_1 = (2.5 \pm 0.3) \text{ cm}^{-1}$ ,  $\Delta\nu_2 = (2.7 \pm 0.3) \text{ cm}^{-1}$ ]:  $m_1 = (19.8 \pm 10) \text{ cm}^{-1}$ ,  $m_2 = (21.3 \pm 10) \text{ cm}^{-1}$ . These estimates are in agreement with the theoretical values:  $m_1 = 22.3 \text{ cm}^{-1}$ ,  $m_2 = 27.3 \text{ cm}^{-1}$ .<sup>7</sup> A good agreement with theory is also observed for the variation of the intensity in the magnetic field (Fig. 2). The discrepancy between the experiment and theory in the region  $H^* < 0.6$  can apparently be attributed to the failure to take into account the interaction of all three oscillations in the region of the fields  $H < 0.6$  T. The experiment makes it possible to estimate  $c_{\text{crit}}$  crudely for the upper crossover:  $2 \times 10^{-4} < c_{\text{crit}_1} < 4 \times 10^{-3}$ , consistent with the theoretical estimates  $c_{\text{crit}_1} \approx 6 \times 10^{-4}$  and  $c_{\text{crit}_2} \approx 2 \times 10^{-3}$ .<sup>7</sup> We shall make more accurate determinations of the values of  $m_1$ ,  $m_2$ ,  $c_{\text{crit}_{1,2}}$ , and of the upper  $\eta$  coefficient after performing an experiment using samples with different, more accurately determined concentrations of manganese.

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