Rearrangement of the spectrum of magnetic excitations of antiferromagnetic CoF_2 with low-concentration MnF_2 impurity

V. V. Eremenko, V. M. Naumenko, S. V. Petrov, and V. V. Pishko

Physicotechnical Institute of Low Temperatures, Academy of Sciences, Ukrainian SSR

(Submitted 28 August 1981)
Zh. Eksp. Teor. Fiz. **82**, 813–826 (March 1982)

With $Co_{1-c}Mn_cF_2$ as an example, the rearrangement of the spectrum of magnetic excitations of a lightly doped antiferromagnet ($c \ll 1$) is investigated in its dependence on the impurity concentration and on the distance between the energy levels of the matrix and of the impurity; this distance is varied with an external magnetic field applied along and perpendicular to the symmetry axis C_4 of the crystal. The results of the experiment are compared with the theory of spin excitations in antiferromagnets with magnetic impurities. The main conclusions of the theory are verified, and the constants of the theory are determined.

PACS numbers: 75.30.Hx, 75.50.Ee

1. INTRODUCTION

This research was devoted to an experimental investigation of the interaction of natural and impurity-induced excitations in lightly doped crystals. The timeliness of such investigations is determined by the great possibilities for use of such crystals in modern technology and by the development of a new theory of lightly doped crystals. The foundations of this theory were laid in the papers of Rashba.[1-3] With the interaction of impurity and exciton states in molecular crystals as an example, it was shown[1] that on approach of the impurity level to the edge of the exciton band, the impurity excitation is delocalized and embraces not only the impurity molecule, but also its surroundings. Here the natural absorption imposes its polarization on the impurity absorption. Such interaction between an impurity molecule and the crystal leads to "ignition" or "extinction" of the impurity absorption line, depending on which edge of the band ($\mathbf{k}=0$ or $\mathbf{k}\neq 0$, respectively) the impurity line is near ($\mathbf{k}$ is the wave vector of the exciton wave).

A second paper[2] developed the theory of impurity absorption near the exciton band in molecular crystals. It obtained the dependence of the position, intensity, and polarization relation of the impurity absorption bands on the distance between the impurity level and the edge of the exciton band. A third paper[3] described the "pumping" of the oscillator strength of the natural oscillations into the impurity oscillations in semiconducting crystals.

The theory of the interaction of impurity and natural excitations received its further development, in particular for antiferromagnets with magnetic impurities, in papers of Ivanov, Loktev, and Pogorelov.[4-7]

Earlier, in accordance with the available experimental data, it had been supposed (see, for example, the reviews[8]) that in order to explain the spectra of magnetic excitations of single crystals of magnetic materials containing a small amount of impurity substitution (with concentration $c \ll 1$), it was not necessary to take into account the interaction of the impurity ions with each other. Then the spectrum of the crystal containing an impurity is described by a simple sum of the spectra of an ideal crystal and of the impurity, in the single-impurity approximation. The results of two experimental papers,[9,10] however, could not be explained within the framework of such a theory. Prokhorov *et al.* (Ref. 9) revealed significant enhancement of the intensity of impurity absorption on approach of the impurity line to the antiferromagnetic resonance (AFMR) line, and Borovik-Romanov *et al.* (Refs. 10) revealed splitting of the absorption lines of AFMR and of impurity resonance instead of intersection of them.

The recently developed theory[4-7] made it possible to explain these experimental data. It was found that allowance for the low-symmetry part of the single-ion anisotropy, in the description of magnetic materials, may lead both to a more complicated type of dependence of AFMR in pure crystals and to a specific resonance interaction of the magnetic excitations of the matrix and of the impurity in lightly doped crystals. This interaction leads to delocalization of the impurity state on approach of the impurity level to the edge of the band of natural states of the crystal. In antiferromagnets with magnetic impurities, with which we shall be concerned hereafter, such an approach is easily accomplished with an external magnetic field, when the impurity level lies below the bottom of the spin-wave band. Delocalization of the wave function of the impurity and the corresponding increase of the radius of the impurity state,

$$r_{\mathrm{imp}}=a[\Omega/(\omega_{\mathrm{AFMR}}-\omega_{\mathrm{imp}})]^{1/2}$$

(here a is the lattice constant, Ω is the coefficient of the quadratic term in the dispersion law near the bottom of the spin-wave band, and ω_{AFMR} and ω_{imp} are the frequencies of the AFMR and impurity absorption lines, respectively), make the oscillations of the matrix and of the impurity interdependent on decrease of the value of $\omega_{\mathrm{AFMR}}-\omega_{\mathrm{imp}}$. This leads to an increase of the intensity of the impurity absorption. And when r_{imp} increases so much that it becomes comparable with the mean distance between impurities, $\bar{r}_{\mathrm{imp}}\sim ac^{-1/3}$, the coupling between the oscillations of separate impurity ions becomes important, and a collective rearrangement of the spectrum of the crystal occurs. Then if $c>c_{\mathrm{cr}}=\eta(m/\Omega)^6$ (η is a numerical factor, m is the constant of

0038-5646/82/030481-09$04.00

resonance interaction of the matrix and the impurity), the rearrangement is a coherent rearrangement (CR): the impurity oscillations form a characteristic band of spin waves, i.e., are described by a wave vector; in the absorption spectrum, a splitting of the lines is observed, accompanied by a pumping of the intensities between them. If $c<c_{cr}$, incoherent rearrangement (ICR) of the spectrum occurs: the neighborhood of the edge of the spin-wave band is found to be filled by fluctuational levels. Then the impurity states do not form a spin-wave band; the concept of an isolated local or quasilocal impurity level also disappears; the absorption lines of the AFMR and of the impurity oscillations fuse.

The purpose of the present work was to investigate experimentally the interaction of natural and impurity-induced magnetic excitations in lightly doped antiferromagnets, and to compare the data obtained with the recently developed theory,[4-7] with tetragonal $Co_{1-c}Mn_cF_2$, $c\ll 1$, as an example. In this crystal, the magnetic moments of the impurity ions Mn^{2+} and of the ions of the Co matrix are collinear, and the energy of the magnetic excitation of the impurity lies below the bottom of the spin-wave band.[11] This makes it possible to investigate the rearrangement of the spectrum of the crystal on approach of the low-frequency AFMR mode from the upper and lower Zeeman sublevels of the magnetic excitation of the impurity, by orientation of the external magnetic field along and perpendicular to the C_4 axis of the crystal. The corresponding ranges of interaction are denoted by the numbers 1, 2, and 3.

2. EXPERIMENTAL METHOD AND SPECIMENS

In the high-frequency range, 22-42 cm^{-1}, the experiment was conducted on a longwave IR diffraction spectrometer.[12] A magnetic field of intensity up to 6 T was produced with a superconducting solenoid. Scanning of the spectrum could be accomplished by sweeping of the frequency at fixed magnetic field or by sweeping the magnetic field at fixed frequency. Both forms of scanning were used in order to increase the accuracy of the experiment on determination of the form and intensity of the absorption lines. The magnetic field was measured with high accuracy (~0.2%) on the basis of the value of the current through the solenoid.

In the frequency range 12–30 cm^{-1}, a pulsed submillimeter spectrometer of transmission type[13] was used. The sources of radiation were backward-wave tubes, the receiver an n-InSb crystal cooled to 4.2 K. The wavelength was determined by means of an interference wavemeter with accuracy 0.2%. The stability of the wavelength depended principally on the stability of the voltage on the decelerating system of the tube (~10^{-4}). By measuring this voltage with a digital voltmeter, it was possible also to determine with high accuracy the wavelength at an arbitrary instant of time, according to a calibration curve taken beforehand.

The specimen was placed at the center of an ordinary solenoid or Helmholtz coil and was clamped by two conical lenses of teflon or quartz.[13] The use of conical lenses and of a spring-controlled mounting made it possible to insure wide-band properties of the spectrometer and small losses during the brining in of radiation to the specimen, and at the same time to choose a pressure on the specimen for which there was no distortion of the resonance lines and the specimens were preserved, even on attainment of the transition field (~22 T) in a CoF_2 crystal, which has a large value of the magnetostriction.

The intensity of the magnetic field was determined by integration of the signal from a test coil, placed in the channel of the pulsed solenoid in the immediate vicinity of the specimen. Calibration of the magnetic field was accomplished on the basis of the EPR line in α-diphenyl-β-picryl hydrazyl (DPPH) or the AFMR line in $RbMnF_3$. A polyethylene pellet containing a small quantity of one of these substances was placed near the specimen. Use of calibration markers, allowance for the nonlinearity of the field-recording channel, and prescription of the zero level of the magnetic field made it possible to improve the accuracy of the field measurement to 1–2%.

In the present work, the determination of the exact value of the intensities of the resonance and impurity lines was of basic importance. Whereas for ordinary (optical) spectrometers this problem has been sufficiently well worked out, for submillimeter spectrometers, operating with pulsed magnetic fields and low-power sources of radiation, with a very irregular amplitude characteristic, there are many difficulties. These difficulties are removed, to a considerable degree, by recording the zero and 100-percent levels of absorption on the screen of a memory oscillograph; they are registered each time immediately before application of the pulsed magnetic field.[13] Thus on the spectrogram, photographed from the oscillograph screen, there are records of the signal and of the zero level of the magnetic field, of the spectrum of the specimen during scanning over the field, and the 0- and 100-percent levels of the signal at $H=0$ (Fig. 1). The presence of all these levels makes it possible to deter-

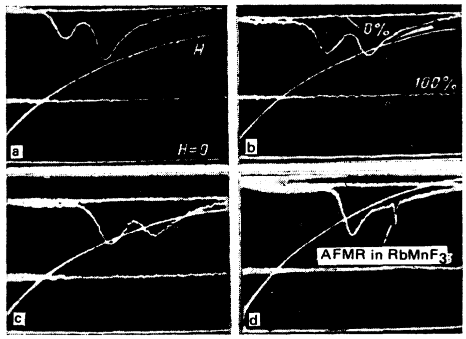

FIG. 1. Oscillograms for $CoF_2+3\cdot 10^{-3}$ Mn^{2+} in interaction region 2. $H\parallel C_4$, specimen thickness 0.07 mm. a) Enhancement of the impurity absorption line on approach of it to the AFMR line, $\nu=22.4$ cm^{-1}. b) Absorption lines near the cross point, $\nu=19.6$ cm^{-1}. c) Absorption lines after the cross point, $\nu=19.1$ cm^{-1}. d) Impurity absorption in the spin-wave band shows up as a distortion of the right wing of the AFMR line, $\nu=15.6$ cm^{-1}.

mine the parameters of the absorption lines with high accuracy.

In the calculation of the absolute values of the integral intensities, corrections were introduced for instrumental functions of the spectrometers and for truncation of the lines. As a rule, the width of the lines exceeded by several times the width of the instrumental function of the spectrometers. Therefore the main correction originated from truncation of the lines. With allowance for these corrections, and also for deterioration of the signal/noise ratio at some frequencies because of a drop in the radiation power, we determine an error in the measurement of the absolute integral intensities of the amount 30%.

In the experiments, single crystals of CoF_2, with MnF_2 impurity concentration from $2\cdot 10^{-5}$ to 10^{-2} by weight, were used. They were grown by the Bridgman method, in platinum crucibles, in an atmosphere of helium evaporated from a Dewar. The MnF_2 impurity was introduced by weight into the mixture for growth of the crystals (CoF_2, melted in a stream of HF). In the crystals obtained, the impurity concentration was monitored by the method of spectral analysis[1)] with calibration of the apparatus on sample powders, prepared from pure CoF_2 and MnF_2, so that the error in the determination of the impurity concentration did not exceed ±20% at $c = 3\cdot 10^{-4}$ and ±5% at $c = 10^{-2}$. Orientation of the single crystals was achieved by x-rays with accuracy ±1°. The specimens were plane-parallel or slightly wedge-shaped plates of size about 3 × 3 mm. Especial attention was paid to the choice of optimum thickness of the specimens, since for correct investigation of the ranges of interaction of the natural and impurity oscillations the absorption lines must not be saturated, while at the same time excessive thinness of the specimens may significantly restrict the range of observation of impurity oscillations. The thickness of the specimens varied from 0.06 to 0.1 mm. Then the intensity of absorption within the interaction range amounted to 50–70%.

The crystals grown showed good conoscope figures, which indicates a high quality of the single crystals; but the AFMR linewidths were quite significant. This made it difficult to determine the intensity of the lines as they approached each other. Therefore the chief measurements were made on specimens cut from a crystal with impurity concentration $4\cdot 10^{-3}$. In them were observed not very broad lines and a quite large splitting at the cross point, the point of equality of the integral intensities of the lines, so that the overlapping of the lines was not great and did not impair the accuracy of determination of the intensity of the lines, while the impurity concentration was sufficient to permit accurate following of the evolution of the impurity and natural absorption lines over a large interval of magnetic fields and frequencies.

Finally, we mention that the use of an ordinary solenoid and of a Helmholtz coil made it possible to investigate the same specimens in three regions of interaction of the impurity and natural lines (Figs. 2 and 4), in which oscillations are excited both with $\mathbf{h}\perp\mathbf{H}$ (in regions 1 and 2) and with $\mathbf{h}\parallel\mathbf{H}$ (region 3), where h is the magnetic vector of the electromagnetic radiation. Of course it was possible to use the Helmholtz coil alone, but we avoided obtaining a field stronger than 13 T in it.

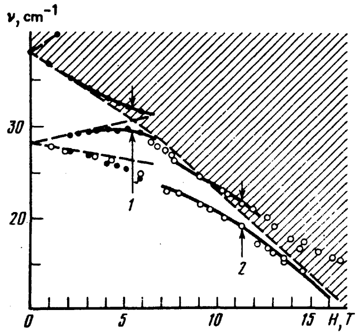

FIG. 2. Frequency-field dependence of the absorption lines in $CoF_2 + 4\cdot 10^{-3}$ Mn^{2+}, $\mathbf{H}\parallel C_4$. Dashed lines: phenomenological calculation for a crystal without impurities. Solid lines: theoretical, for frequency scanning of the spectrum.[7] The dark points were obtained with frequency scanning of the spectrum, the light points with field scanning. The spin-wave band is hatched. 1) $\Delta\nu_1$, interaction range 1; 2) $\Delta\nu_2$, interaction range range 2.

3. THEORY

A theoretical treatment of the rearrangement of the spectrum of magnetic excitations of CoF_2 with MnF_2 impurity in an external magnetic field was carried out by Ivanov, Loktev, and Pogorelov.[5-7] We shall discuss the results of the theory that have a relation to our experiment.

As was shown,[5] the Hamiltonian of a crystal containing impurities can be represented in the form

$$\mathscr{H}=\mathscr{H}_{Co}+\mathscr{H}'; \quad \mathscr{H}_{Co}=\sum_{\mathbf{k};j=1,2}[\omega(\mathbf{k})+(-1)^{j-1}\mu gH]\beta_j^+(\mathbf{k})\beta_j^-(\mathbf{k}),$$

$$\mathscr{H}'=\frac{1}{Z}\sum_{\mathbf{p}_\alpha,\rho}\{J'\sigma_{\mathbf{p}_\alpha}\mathbf{S}_{\mathbf{p}_\alpha+\rho}-J_\perp(S^x_{\mathbf{p}_\alpha}S^x_{\mathbf{p}_\alpha+\rho}+S^y_{\mathbf{p}_\alpha}S^y_{\mathbf{p}_\alpha+\rho})-J_\parallel S^z_{\mathbf{p}_\alpha}S^z_{\mathbf{p}_\alpha+\rho}$$
$$+A(S^z_{\mathbf{p}_\alpha})^2-B_\alpha[(S^x_{\mathbf{p}_\alpha})^2-(S^y_{\mathbf{p}_\alpha})^2]+\mu g_{Co}HS^z_{\mathbf{p}_\alpha}-2\mu H\sigma^z_{\mathbf{p}_\alpha}\}. \qquad (1)$$

We are considering the case of z orientation of the external magnetic field: $\mathbf{H}\parallel C_4$; C_4 is the fourfold symmetry axis of the crystal. Here $\mathscr{H}_{Co}$ is the part of the Hamiltonian that is exactly diagonalizable by means of the Bogolyubov-Tyablikov transformation; it corresponds to pure CoF_2 in magnetic fields H small in comparison with the flip field of the sublattices (approximately 22 T[14]):

$$\omega(k)=\omega(0)+(4\pi)^{-1/3}\Omega(ak)^2 \qquad (2)$$

is the dispersion law near the bottom of the magnon band of CoF_2; $\mathscr{H}'$ is the impurity part of the Hamiltonian, written on the assumption that the ions with spin $\sigma = 5/2$ are distributed with uniform probability $c \ll 1$ over sites $\mathbf{p}_\alpha$ ($\alpha = 1, 2$ is the number of the magnetic sublattice); the g factor of the Mn^{2+} ions is taken equal to 2, and their exchange interaction ($J' > 0$) with neighboring Co^{2+} ions is taken to be isotropic; $S_{\mathbf{p}_\alpha+\rho}$ and $\sigma_{\mathbf{p}_\alpha}$ are the spin operators of the matrix and of the impurity, respectively; $A, B > 0$ are the parameters of the single-ion anisotropy; Z is the number of nearest neighbors.

In the present case ($Co_{1-c}Mn_cF_2, \mathbf{H} \parallel C_4$), we may neglect interaction between impurity ions belonging to different sublattices. When the eigenfrequencies of the oscillations of the impurity ions are close at $H=0$, the interaction between them is exponentially small: $\sim \exp(-\bar{r}_{imp}/r_{imp}), \bar{r}_{imp} \gg r_{imp}$. And when, with increase of H, one of the impurity levels approaches the edge of the spin-wave band and the radius of its state increases, the interaction between impurities with opposite directions of the spins gives only an insignificant nonresonant correction to the frequencies of their oscillations. Therefore further consideration of the natural and impurity oscillations may be carried out for each Zeeman impurity sublevel independently, since each of these sublevels belongs to ions of one of the sublattices.

The spectrum of eigenfrequencies of the crystal and the absorption spectrum of electromagnetic radiation can be described by means of a diagonal Green function $\hat{G}(\mathbf{k},\mathbf{k})$. For the upper Zeeman sublevel of the impurity (region 1 of interaction in Fig. 2), in the approximation linear with respect to c, one can obtain the following expression for $G(\mathbf{k},\mathbf{k})$:

$$\hat{G}(\mathbf{k},\mathbf{k})=[\hat{G}^{(0)-1}(\mathbf{k},\mathbf{k})-\hat{R}_{\mathbf{k}}]^{-1};$$

$$\hat{G}^{(0)-1}(\mathbf{k},\mathbf{k})=-\begin{pmatrix} (\hat{g}_{\mathbf{k}}^{+})^{-1} & 0 \\ 0 & (\hat{g}_{\mathbf{k}}^{-})^{-1} \end{pmatrix}.$$

$$(\hat{g}_{\mathbf{k}}^{\pm})^{-1}=\begin{pmatrix} \varepsilon-\omega\pm\mu gH & M_2\gamma_{\mathbf{k}} \\ M_2\gamma_{\mathbf{k}} & \varepsilon+\omega\mp\mu gH \end{pmatrix}, \tag{3}$$

$$M_2=M_{20}-\varkappa(\mu H)^2, \quad M_{20}=J_{\perp}(x^2-y^2)/2, \quad \varkappa=J_{\perp}(x'^2-y'^2)/2,$$

$$\varepsilon=J_{\parallel}s_0+[(J_{\parallel}s_0+A)^2+3B^2]^{1/2}-[(J_{\parallel}s_0-A)^2+3B^2]^{1/2},$$

$$\gamma_{\mathbf{k}}=\frac{1}{Z}\sum_{\rho} e^{i\mathbf{k}\rho}.$$

Here the quantities x, v, x', and v' are defined in Ref. 7, $J_{\perp}$ and $J_{\parallel}$ are exchange constants, and s_0 is the root of the equation

$$(s_0-1/2)^2[(s_0+A/J_{\parallel})^2+(\sqrt{3}B/J_{\parallel})^2]=(s_0+A/J_{\parallel})^2.$$

In the expression for the matrix of the polarization operator

$$\hat{R}_{\mathbf{k}}=c\psi^{+}(\mathbf{k})\hat{V}(\hat{1}-\hat{G}^0\hat{V})^{-1}\psi(\mathbf{k}). \tag{4}$$

$\hat{V}$ is the matrix of the perturbation produced by the impurity center;

$$\hat{G}^0=\frac{1}{N}\sum_{\mathbf{k}'\neq\mathbf{k}}\psi(\mathbf{k}')\hat{G}^{(0)}(\mathbf{k}',\mathbf{k})\psi^{+}(\mathbf{k}');$$

$\psi(\mathbf{k}')$ and $\psi^{+}(\mathbf{k})$ are the column and row vectors in the space of the S representation of the point group of the impurity center[15]; and N is the number of sites in the crystal.

The absorption spectrum far from the region of collective rearrangement

The poles of the matrix $G(\mathbf{k},\mathbf{k})$, which determine the spectrum of elementary excitations in a crystal with impurities, are the solutions of the equation

$$D_{\mathbf{k}}=\det[\hat{G}^{(0)-1}(\mathbf{k},\mathbf{k})-\hat{R}_{\mathbf{k}}]=0. \tag{5}$$

The eigenfrequency $\omega_0^{+}(H)$ (the index + everywhere corresponds to the upper Zeeman sublevel of the impurity), in the single-impurity approximation, is determined by the root of the equation

$$D^{+}(\omega_0^{+}(H), H)=0, \tag{6}$$

where

$$D^{+}(\omega,H)=1-V^{+}f^{+}(\omega,H),$$

$$V^{+}=M_2^2+(\varepsilon_1+\omega)V_2+V_3^2(\varepsilon_1^2-\omega^2)f^{-}(\omega.H)(W^{+})^{-1},$$

$$W^{+}=1-[M_2^2+V_1(\varepsilon_1-\omega)]f^{-}(\omega,H),$$

$$f^{\pm}(\omega,H)=\frac{1}{N}\sum_{\mathbf{k}}\gamma_{\mathbf{k}}^2[\Delta^{\pm}(\mathbf{k})]^{-1},$$

$$\Delta^{\pm}(\mathbf{k})=(\omega\pm\mu gH)^2-\omega^2(\mathbf{k});$$

V_1, V_2, V_3 are the elements of the perturbation matrix $\hat{V}$.

In the single-impurity approximation and with consideration only of the upper Zeeman sublevel of the impurity, the expression (5) can be written in the form

$$D_{\mathbf{k}}=\Delta^{-}(\mathbf{k})[\Delta^{+}(\mathbf{k})-R^{+}(\mathbf{k})]=0, \quad R^{+}(\mathbf{k})=c\gamma_{\mathbf{k}}^2V^{+}[D^{+}(\omega,H)]^{-1}. \tag{7}$$

The first factor in (7) corresponds to the unperturbed upper branch $\omega_1(\mathbf{k},H)$, while the second gives the dispersion equation for oscillations of the lower spin-wave branch $\omega_2(\mathbf{k},H)$ of the matrix coupled with oscillations of the impurity.

The states of the elementary excitations in a crystal with impurities can be of two types: band states (characterized by a wave vector) or localized (at one or several impurity sites) states. The spectrum of band states, corresponding to coupled oscillations of the lower spin-wave branch of the matrix and oscillations of the impurity, is determined by the solution of the dispersion equation

$$(\omega+\mu gH)^2-\omega^2(\mathbf{k})-\mathrm{Re}\,R^{+}(\mathbf{k})=0. \tag{8}$$

Such solutions, however, have physical meaning only if they lie within the range of convergence of the renormalized representation for $R^{+}(\mathbf{k})$.[16] The region outside the spectrum of band states is filled by fluctuational localized states; outside the region of concentrational broadening of the impurity level $\Delta^{+}(H)$, the density of such states is provided mainly by states localized at pairs of impurities lying close together. When the impurity level $\omega_0^{+}(H)$ is far from the region of collective rearrangement of the spectrum, so that $r_{imp}(H) \ll \bar{r}_{imp}$, solutions of Eq. (8) lying outside of $\Delta^{+}(H)$ do not correspond to real band states, since the renormalized representation for $R^{+}(k)$ in this region diverges. Thus far from the region of collective rearrangement of the spectrum, states near the impurity level remain localized.

It can be shown that in small fields H, the field dependence of the impurity frequency is linear:

$$\omega_0^{+}(H)=\omega_0^{+}(0)+\mu g_{eff}H.$$

With increase of field, the $\omega_0^{+}(H)$ dependence becomes significantly nonlinear, as is described by numerical solution of equation (6). The impurity level begins to be "repelled" by the bottom of the spin-wave band of the crystal (see Fig. 2).

The integral (integrated over ω at $H=$ const) intensity of absorption by the impurity level $\omega_0^{+}(H)$ can, with the aid of the Green function (3), be put into the form

$$I_{\text{imp}}(H)=cK\Lambda(H)P(H),\quad P(H)=\{1+{}^1/_2[\delta_+d_+^{-1}(H)]^{1/2}\}^{-1},$$
$$\Lambda(H)=[1+m_+d_+^{-1}(H)]^2,\quad d_+(H)=\omega_2(0,H)-\omega_0^+(H),\qquad(9)$$

where m_+ is the constant of resonance interaction of the impurity with the matrix, $\delta_+=m_+^4/\Omega^3$, and K is a numerical factor.

As has been shown,[4] the quantity $P(H)$ is the square of the modulus of the wave function ψ of the impurity state at the impurity site. Far from the region of rearrangement of the spectrum, ψ is practically completely localized at the impurity site, and $P(H)=1$. With approach to the region of rearrangement, the state described by ψ is delocalized, and $P(H)$ decreases. But as is evident from (9), there is still the factor $\Lambda(H)$, which increases faster than $P(H)$ decreases. The increase of $\Lambda(H)$ is due to the fact that a contribution to the absorption at the impurity frequency is made not only by the oscillations of strictly impurity type, but also by oscillations, coupled with them through the constant m_+, of the matrix with $k=0$, and this corresponds to an increase of r_{imp}. The resonance increase of $\Lambda(H)$ leads to "ignition" of the impurity absorption line when it approaches the AFMR line, at the cost of weakening of the latter. It must be noted that when the impurity level and the optically inactive (in our case, the upper) edge of the band approach each other, $\Lambda(H)$ will not undergo a resonance increase, while at the same time ψ will be, as before, delocalized. As a result, instead of ignition, "extinction" of the impurity line will occur.

Collective rearrangement of the spectrum

When H reaches the value H_r^+ at which $\bar{r}_{\text{imp}}\sim r_{\text{imp}}$, the interaction between separate impurity ions becomes significant, collective rearrangement of the spectrum occurs, and the single-impurity approximation ceases to be valid.[4] Here the broadening $\Delta^+(H)$ of the impurity level increases from an exponentially value to the value

$$\Delta^+(H_r^+)\sim c^{1/2}\frac{m_+^2}{\Omega}\frac{1}{c^{1/2}+(c_{\text{cr}}^+)^{1/2}},\quad c_{\text{cr}}^+=\eta\left(\frac{m_+}{\Omega}\right)^6.$$

The collective rearrangement of the spectrum may be coherent or incoherent, depending on the relation between c and c_{cr}^+. For $c\ll c_{\text{cr}}$, the value of

$$\Delta^+(H_r^+)\sim c^2\,\Omega$$

is of the same order of magnitude as

$$d_+(H_r^+)=\omega_2(0,H_r^+)-\omega_0^+(H_r^+),$$

so that for $H\approx H_r^+$ the impurity absorption line fuses with the AFMR line. The intensities of the two lines are comparable. A neighborhood of the edge of the spin-wave band, of width $\sim c^{2/3}\Omega$, is filled with fluctuational levels, and here one may not speak of band states, but the concept of an isolated local or quasilocal impurity level also disappears. Such structure of the spectrum corresponds to incoherent resonance (ICR). With increase of the field intensity, the ICR disappears when again $r_{\text{imp}}(H)<\bar{r}_{\text{imp}}$. But here the impurity level is not of resonance type but virtual, because of the large attenuation caused by transitions of impurity excitations to excitations of the spin-wave spectrum of the crystal.

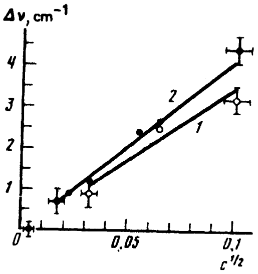

FIG. 3. Dependence of the value of the frequency interval between absorption lines, repelling each other at the cross point, on impurity concentration. 1) $\Delta\nu_1$ in region 1; 2) $\Delta\nu_2$ in region 2.

On further increase of the field, there appears in the continuous spectrum a quasilocal resonance impurity level.

If $c\gg c_{\text{cr}}$, then at fields $H\approx H_r^+$ a coherent-resonance (CR) spectrum occurs. For CR, in contrast to ICR, solutions of the dispersion equation (8) written for the case $H\sim H_r^+$ have physical meaning also within a certain neighborhood of the impurity level (H_r^+ is the field for fusion of the impurity line and AFMR in ICR in the limit $c\to 0$). The solutions of this equation at $\mathbf{k}=0$ have the form

$$\Omega_{1,2}(H)=\omega_r^++\mu\frac{g-g_r^+}{2}(H_r^+-H)\pm\left\{\left[\mu\frac{g+g_r^+}{2}(H-H_r^+)\right]^2+cm_+^2\right\}^{1/2},$$
$$\omega_r^+=\omega_0^+(H_r^+),\quad g_r^+=-\frac{(\partial V^+/\partial H)_{H_r^+}}{\mu(\partial V^+/\partial\omega)_{\omega_r^+}}.\qquad(10)$$

The results of a calculation of the absorption frequencies shown in Fig. 2 were obtained, in contrast to the single-impurity approximation (8), by means of a certain self-consistent approximation similar to the method of the coherent potential, and therefore are correct in the region of collective rearrangement of the spectrum.

In fields $H>H_r^+$, a second dispersion-impurity branch of the spectrum, Ω_2, is formed, of width

$$\omega_0(H)-\Omega_2(H)>\Delta^+(H)\sim c^{1/2}m_+^2/\Omega.$$

Furthermore, for $k<k_{\max}\sim c^{1/3}a^{-1}$ the attenuation of impurity states over a wavelength is small, so that impurity band states can actually exist. The width of the quasigap between the ground and impurity bands, $\omega_2(0)-\omega_r^+$, amounts to $c^{\frac{1}{2}}m_+$ (Ref. 4), so that the distance between the AFMR frequency $\Omega_1(H)$ and the impurity resonance frequency $\Omega_2(H)$ amounts to $2c^{\frac{1}{2}}m_+$. The experimental $\Delta\nu(c)$ relation, corresponding to $2c^{\frac{1}{2}}m_+$, is shown in Fig. 3. The value of $2c^{\frac{1}{2}}m_+$ exceeds the value of $\Delta^+(H)$; therefore in CR, two lines are observed in the absorption spectrum over the whole frequency range; this corresponds to splitting of the absorption lines. For $H>H_r^+$, the impurity oscillation falls within the spin-wave spectrum of the crystal, in which a narrow region (quasigap) is formed where all states are localized and where pairs of impurities are produced, located at distances $r_{\text{cr}}<\bar{r}_{\text{cr}}$. When the width of the quasigap becomes comparable with $\Delta^+(H)$ (on further increase of H), the quasigap "collapses," and there remains in the spectrum a single continuous region of band states with a quasilocalized level, similar to the case of ICR.

The intensities of the two absorption lines with frequencies $\Omega_1(H)$ and $\Omega_2(H)$, in the CR region, have the relation

$$\frac{I_1}{I_2}=\frac{\Omega_1(H)-\omega_0^+(H)}{\omega_0^+(H)-\Omega_2(H)}. \tag{11}$$

whence it is evident that with increase of H, there occurs in this region a "pumping" of intensity from the AFMR line $[\Omega_1(H)]$ into the impurity line $[\Omega_2(H)]$ (see Fig. 1). The results of a calculation by formula (11) for the value $c=3\cdot 10^{-4}$ are shown in Fig. 4.

As is easy to see, the treatment of the interaction of the low-frequency AFMR mode with the lower Zeeman sublevel of the impurity oscillation (interaction region 2 in Fig. 2) is completely equivalent to that given above for the upper Zeeman impurity sublevel with the field value H replaced by $-H$. Then the dispersion equation (7), for example, will have the form

$$\Delta^+(\mathbf{k})\,[\Delta^-(\mathbf{k})-R^-(\mathbf{k})]=0,$$
$$R^-(\mathbf{k})=c\gamma_{\mathbf{k}}^2V^-[D^-(\omega,H)]^{-1},\quad D^-(\omega,H)=1-V^-f^+(\omega,H),$$
$$V^-=M_2^2-(\varepsilon_2-\omega)V_1+V_3^2(\varepsilon_2^2-\omega^2)f(\omega,H)(W^-)^{-1}.$$
$$W^-=1-[M_2^2+V_2(\varepsilon_2-\omega)]f^-(\omega,H).$$

Similarly, all the expressions describing the upper impurity sublevel and its interaction with AFMR remain in force for the lower level if in them we replace ω_r^+, m_+, and V^+ by ω_r^-, m_-, and V^- respectively.

The results of a theoretical treatment of the interaction of oscillations of the impurity with the low-frequency AFMR mode in CoF_2+cMn^{2+} (everywhere hereafter, we shall for simplicity use the notation CoF_2+cMn^{2+} instead of $Co_{1-c}Mn_cF_2$) for the case $\mathbf{H}\perp C_4$ (interaction region 3 in Fig. 4) were given in Ref. 18. Calculation of the frequency-field relation of the absorption lines (see Fig. 4) shows that even for orientation $\mathbf{H}\perp C_4$, splitting of the absorption lines occurs in CR. In ICR, the absorption lines must fuse.

4. RESULTS OF THE EXPERIMENT AND COMPARISON WITH THEORY

Frequency-field relation of the absorption lines ($\mathbf{H}\parallel C_4$)

The field dependence of the frequency of the natural and impurity absorption lines in $CoF_2+4\cdot 10^{-3}Mn^{2+}$, for the case $\mathbf{H}\parallel C_4$, were investigated over the frequency range 12–42 cm^{-1}, in constant magnetic fields up to 6 T and in pulsed fields up to 17 T. At $H=0$, the widths of the impurity and AFMR lines were about 1.5 and 2.2 cm^{-1}, respectively, and they did not change appreciably with change of H. The shape of the lines was intermediate between Lorentzian and Gaussian (see also Ref. 19). The experimental points are shown in Fig. 2. Theoretical curves were calculated by use of the values $A=24$ cm^{-1}, $B=43$ cm^{-1}, $g_{Co}=2.6$.[7,20] The parameters $J_\parallel=35$ cm^{-1} and $J_\perp=42.1$ cm^{-1} were so chosen as to insure best coincidence of the theoretical and experimental data. The good agreement of the experimental and theoretical relations confirms the correctness of the theoretical model chosen for description of the system CoF_2+cMn^{2+}.

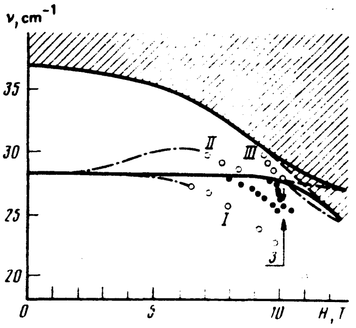

FIG. 4. Frequency-field relation of absorption lines in CoF_2+cMn^{2+}, $\mathbf{H}\perp C_4$. The experimental points were obtained by scanning of the spectrum with respect to field. Light points, $c=3\cdot 10^{-3}$; dark, $c=3\cdot 10^{-4}$. Solid and dashed lines, calculation for $c=2\cdot 10^{-4}$, respectively with and without allowance for CR.[18] Dashed-dotted curve and experimental points continuing it, proposed behavior of the oscillations for $c=3\cdot 10^{-3}$. The spin-wave band is hatched; 3) $\Delta\nu_3$, interaction region 3.

Interaction region 1

Interaction region 1 was investigated in specimens of CoF_2 having concentrations of Mn^{2+} impurity in the amounts $c=10^{-3}$, $4\cdot 10^{-3}$, and 10^{-2} by weight. For all three concentrations, the rearrangement of the spectrum was coherent: on approach to the AFMR line, the impurity line was enhanced, in the rearrangement region splitting of the absorption lines was observed, and it was accompanied by pumping of the intensity from one line to another. When the impurity line fell within the spin-wave band, it rapidly diminished in intensity and broadened. The variation of the value $\Delta\nu$ of the splitting of the absorption lines at the cross point (the point of equality of the integral intensities of the lines) with the concentration of the impurity is shown in Fig. 3. Within the limits of experimental error, the dependence of $\Delta\nu$ on $c^{\frac{1}{2}}$ is linear, in accordance with the theoretical relation.

Figure 5 shows the ratio of the integral intensity of the impurity absorption line to the intensity of the AMFR line in interaction regions 1 and 2 for impurity concentration $c=4\cdot 10^{-3}$. The theoretical variation of

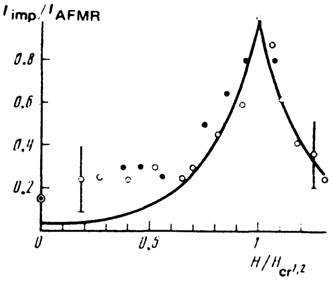

FIG. 5. Variation of the ratio of integral intensities of absorption lines in $CoF_2+4\cdot 10^{-3}$ Mn^{2+}. Dark points, for interaction region 1; light, for interaction region 2. Solid curve, theory.[7] The intensities of the absorption lines of AFMR and of the impurity at $H=0$ are $2I_{AFMR}=600$ cm^{-1} and $2I_{imp}=100$ cm^{-1} (the factor 2 is due to the presence of degeneracy of the levels at $H=0$). $H_{cr1,2}$ are the fields of the cross points in regions 1 and 2.

the ratio of integral intensities is plotted according to formula (11). The disagreement of experiment and theory in the region $H/H_{cr1,2} < 0.5$ confirms the inapplicability of this formula far from the cross point. It may also indicate the necessity for taking account of all three oscillations in this field range.

Interaction region 2

Interaction region 2 was investigated on specimens of CoF_2 containing Mn^{2+} impurity in the amounts $c = 2\cdot 10^{-5}$, $3\cdot 10^{-4}$, $5\cdot 10^{-4}$, 10^{-3}, $3\cdot 10^{-3}$, $4\cdot 10^{-3}$, and 10^{-2} by weight. In the specimen containing $2\cdot 10^{-5}$ Mn^{2+} impurity, the rearrangement of the spectrum was incoherent. The difference of ICR from CR showed up in the fact that the impurity absorption line fused with the AFMR line and that its width did not change appreciably during this process, whereas for CR the widths of the lines had a tendency to become equal. After fusion of the lines, the impurity line never manifested itself within the spin-wave band of characteristic excitations of the crystal. For the remaining specimens, coherent rearrangement of the spectrum was observed, similar to that described for region 1. The values of the linewidths of the the impurity absorption and of AFMR at frequency 22.5 cm^{-1} (far from the cross point), as well as the coordinates of the cross points, are given as functions of the impurity concentration in Table I. It is seen that with increase of concentration the cross point shifts toward larger ν (smaller H); this is caused by the beginning of the rearrangement of the spectrum at smaller fields (at smaller r_{imp}) because of decrease of $\bar{r}_{imp}$.

The variation of the amount of splitting of the absorption lines at the cross point with the impurity concentration is shown in Fig. 3. For interaction region 2, as well as for region 1, the variation of $\Delta\nu$ with $c^{\frac{1}{2}}$ is linear; this also corroborates the correctness of the theoretical description given.

Interaction region 3

In the orientation $H \perp C_4$, specimens were investigated with impurity concentration $c = 3\cdot 10^{-4}$, 10^{-3}, $3\cdot 10^{-3}$, and 10^{-2}. For these concentrations, the rearrangement of the spectrum was found to be coherent, with interaction of the lower AFMR mode with the lower impurity oscillation. For concentrations $3\cdot 10^{-4}$ and 10^{-3}, the amounts of splitting were 0.7 and 1.5 cm^{-1}, respectively; this agrees with the value 1 cm^{-1} for $c = 2\cdot 10^{-4}$ (Ref. 18) and is close to the value of the splitting in region 2.

TABLE I. Linewidth of AFMR and of the impurity and coordinates of the cross point in interaction region 2.

c	$2\cdot 10^{-5}$	$3\cdot 10^{-4}$	$5\cdot 10^{-4}$	10^{-3}	$3\cdot 10^{-3}$	$4\cdot 10^{-3}$	10^{-2}
Δ_{AFMR}, cm^{-1}	1.0	1.3	1.2	1.2	1,6	2,2	2,4
Δ_{imp}, cm^{-1}	0.2	0.5	0.5	1.0	1.6	1.5	3.2
$\nu_{cr\,2}$, cm^{-1}	17 (fusion point)	18.3	18.1	19	20	20.5	21.7
$H_{cr\,2}$, T	13.5 (fusion point)	12.9	13.2	12.6	11.8	11.1	10.2

But for concentrations $3\cdot 10^{-3}$ and 10^{-2}, the amounts of cross splitting were 6 and 9 cm^{-1} respectively; these considerably exceed the values predicted by theory (2.5 and 4.2 cm^{-1}). This disagreement can be explained by the presence of still another absorption line in the region of rearrangement of the spectrum. We actually detected this line in the specimen with impurity concentration $3\cdot 10^{-3}$ and identified it with the upper impurity oscillation (Fig. 4, oscillation II). The dashed-dotted lines in this figure give the approximate course of the frequency-field relations of oscillations I, II, and III. In this crystal, the widths of the absorption lines are appreciable, but the slopes of the field dependences of the frequencies of the impurity oscillations (and, near the cross, also of the AFMR) are small (for $H \perp C_4$); and in consequence of the redistribution of intensity between the oscillations during sweep of the spectrum by the magnetic field, the maxima of the observed absorption lines are found to be shifted from their natural positions. The absorption lines of all three oscillations overlap each other; therefore it is difficult to determine accurately the redistribution of intensities among them. So far we have succeeded only in determining that the cross field of oscillations II and III approximately coincides with the cross field of oscillations I III (apparently the first of these lies slightly higher in frequency). Hence we understand the so strong influence of the first of these oscillations on the second upon increase of the impurity concentration.

Thus in a quantitative description of the process of collective rearrangement of the spectrum in region 3, it is necessary to take into account the interaction of all three oscillations, and the theory[4-7] briefly set forth in Sec. 3 is in need of modification. But the key theoretical conclusions—the possibility of collective rearrangement of the spectrum, the splitting of absorption lines during CR, the presence of a critical concentration, etc.—remain in force.

We note also that since the absorption line II owes its large intensity to interaction with oscillation III (AFMR), its polarization also corresponds to the polarization of AFMR ($\mathbf{h} \parallel \mathbf{H}$, see Refs. 1 and 2), and not $\mathbf{h} \perp \mathbf{H}$, as should be the case outside the region of collective rearrangement of the spectrum.

The new absorption line II was observed in the specimen with impurity concentration $c = 3\cdot 10^{-3}$. It could not be observed in the other specimens. Here the thickness of all the specimens was 0.07–0.1 mm. In the specimen with $c = 10^{-2}$, the "absence" of the line is apparently explained by the significant linewidth (see Table I). In the specimens with $c = 3\cdot 10^{-4}$ and 10^{-3}, the intensity of this line should be weak at the given specimen thickness, so that it would be difficult to observe. Far from the region of rearrangement of the spectrum, the line under consideration is no longer enhanced by interaction with the AFMR line; this explains the absence of experimental points on the rising section of the frequency-field relation of oscillation II. Furthermore, a broad line is here to be expected on sweeping with the field, because of the slight dependence of the resonance frequency on the field.

In our view, the absorption line corresponding to oscillation II was present also in the spectrograms of Fig. 36 of Ref. 11, which investigated the interaction of impurity and natural magnetic oscillations in $CoF_2 + 2\cdot 10^{-4}\ Mn^{2+}$. There this line showed up as a kink on the right wing of the absorption line of the lower impurity oscillation I. Its observation at small impurity concentration proved possible because of the use of thick specimens (0.5–1.5 mm).

Quantitative comparison with theory

The carrying out of the experiment on specimens with different impurity contents makes it possible substantially to increase the accuracy of determination of the constants of resonance interaction m_+ and m_- and of the critical concentrations c_{cr}^+ and c_{cr}^- for interaction regions 1 and 2 respectively, and also to determine the numerical factor η in the expression for c_{cr}. For interaction region 2, ICR is observed at impurity concentration $c = 2\cdot 10^{-5}$, and CR is observed at $c = 3\cdot 10^{-4}$; consequently, the critical concentration lies within these limits, $2\cdot 10^{-5} < c_{cr}^- < 3\cdot 10^{-4}$, whence the most probable value is $c_{cr}^- = (1.5 \pm 1)\cdot 10^{-4}$. The slope of the straight lines in Fig. 3 corresponds to the values $m_+ = 17.5 \pm 2$ cm^{-1}, $m_- = 20.5 \pm 1.5$ cm^{-1}, found from the expression $\Delta\nu_{1,2} = 2m_\pm c^{\frac{1}{2}}$.

Use of the value of the quantity $\Omega = 45 \pm 5$ cm^{-1}, calculated from results of a neutron-diffraction analysis of the dispersion of spin waves in CoF_2 (Ref. 21), enables us to determine the numerical factor η by the formula $c_{cr}^\pm = \eta(m_\pm/\Omega)^6$, starting solely from experimental data. Taking the numerical values of c_{cr}^- and m_- for region 2, we get $\eta = 0.02$, which agrees well with the theoretical value $\eta = 0.05$, since allowance for the experimental errors gives a permissible value 0.08. Use for interaction region 1 of the value $\eta = 0.02$ and of $\Omega = 45$ cm^{-1} and $m_+ = 17.5$ cm^{-1} gives the value $c_{cr}^+ = 7\cdot 10^{-5}$.

Table II, for comparison, gives the values of the constants obtained in the present work and the theoretical values.[7] It is seen that the experimental and theoretical values of the constants of resonance interaction m_+ and m_- and of the factor η agree satisfactorily. The difference of the theoretical values of c_{cr}^+ from the experimental is due to the high power to which the constants $m_\pm$ are raised in the formula for $c_{cr}^\pm$.

5. CONCLUSION

Cobalt fluoride with manganese impurity was found to be a convenient object for investigation of the influence of an impurity on the energy spectrum of a crystal. The investigation carried out confirms the deductions of the theory to the effect that the presence and character of the rearrangement of the spectrum are determined by the concentration of the impurity and by the distance between the energy levels of the matrix and of the impurity. Still unconfirmed so far is the deduction of the theory regarding formation of an impurity magnon band during coherent rearrangement of the spectrum. We hope that experiments we plan on two-magnon absorption will shed some light on this question. Experiments on scattering of neutrons would make possible a direct test of this deduction, but one must remember the necessity for high accuracy in such an experiment: even at impurity concentration 1%, the width of the impurity band is estimated to be of amount 1–2 cm^{-1}, which is difficult for neutron spectroscopy.

TABLE II. Experimental and theoretical values of the constants of collective interaction for $Co_{1-c}Mn_cF_2$.

	m_+, cm^{-1}	m_-, cm^{-1}	c_{cr}^+	c_{cr}^-	η	Ω, cm^{-1}
Experiment	17.5	20.5	$7\cdot 10^{-5}$	$1.5\cdot 10^{-4}$	0.02	45 (Ref. 21)
Theory[7]	22.3	27.3	$6\cdot 10^{-4}$	$2\cdot 10^{-3}$	0.05	46.5

The results of the investigation have also shown that in a number of cases one may not use a two-level system. More complicated theoretical calculations are necessary.

[1)] We thank L. E. Usenko for his careful carrying out of the analysis.

---

[1] É. I. Rashba, Opt. Spektrosk. **2**, 568 (1957).

[2] É. I. Rashba, Fiz. Tverd. Tela (Leningrad) **4**, 3301 (1962) [Sov. Phys. Solid State **4**, 2417 (1963)].

[3] É. I. Rashba and G. É. Gurgenishvili, Fiz. Tverd. Tela (Leningrad) **4**, 1029 (1962) [Sov. Phys. Solid State **4**, 759 (1962)].

[4] M. A. Ivanov and Yu. G. Pogorelov, Zh. Eksp. Teor. Fiz. **76**, 1010 (1979) [Sov. Phys. JETP **49**, 510 (1979)].

[5] V. M. Loktev and Yu. G. Pogorelov, Fiz. Nizk. Temp. **5**, 483 (1979) [Sov. J. Low Temp. Phys. **5**, 233 (1979)].

[6] M. A. Ivanov, Yu. G. Pogorelov, and V. M. Loktev, Spinovye vozbuzhdeniya v antiferromagnitnom CoF_2 pri vysokoĭ kontsentratsii primesi Mn^{2+} (Spin Excitations in Antiferromagnetic CoF_2 at High Concentration of Mn^{2+} Impurity). All-Union Conference on the Physics of Low Temperatures, 23–26 January 1979. Abstracts of Papers, Moscow, 1978, p. 24.

[7] M. A. Ivanov, V. M. Loktev, and Yu. G. Pogorelov, Perestroĭka spektra spinovykh vozbuzhdeniĭ anizotropnogo antiferromagnetika s magnitnymi primesyami vo vneshnem pole (Rearrangement of the Spectrum of Spin Excitations of an Anisotropic Antiferromagnet with Magnetic Impurities in an External Field). Preprint ITF-80-40R, Kiev, 1980; Fiz. Nizk. Temp. **7**, 1401 (1981) [Sov. J. Low Temp. Phys. **7**, 679 (1981)].

[8] R. A. Cowley and W. J. L. Buyers, Rev. Mod. Phys. **44**, 406 (1972); R. Weber, Z. Phys. **213**, 299 (1969).

[9] A. S. Prokhorov and E. G. Rudashevskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. **22**, 214 (1975) [JETP Lett. **22**, 99 (1975)].

[10] A. S. Borovik-Romanov and V. F. Meshcheryakov, Pis'ma Zh. Eksp. Teor. Fiz. **8**, 425 (1968) [JETP Lett. **8**, 262 (1968)]; B. S. Dumesh, V. M. Egorof, and V. F. Meshcheryakov, Zh. Eksp. Teor. Fiz. **61**, 320 (1971) [Sov. Phys. JETP **34**, 168 (1972)].

[11] K. Kocharyan, Dissertation in competition for the degree of candidate of physical-mathematical sciences, Institute of Physical Problems, Academy of Sciences, USSR, 1973; B. Enders, P. L. Richards, W. E. Tennant, and E. Catalano, AIP Conf. Proc. **10**, 179 (1973).

[12] V. M. Naumenko, V. I. Fomin, and V. V. Eremenko, Prib. Tekh. Eksp. No. 5, 223 (1967) [Instrum. Exp. Tech. No. 5,

1201 (1967)]; V. M. Gredescul, S. A. Gredescul, V. V. Eremenko, and V. M. Naumenko, J. Phys. Chem. Solids **33**, 859 (1972).

[13] V. M. Naumenko, V. V. Eremenko, and A. V. Klochko, Prib. Tekh. Eksp. No. 4, 159 (1981).

[14] Yu. M. Gufan, K. H. Kocharyan, A. S. Prokhorov, and E. G. Rudashevskiĭ, Zh. Eksp. Teor. Fiz. **66**, 1155 (1974) [Sov. Phys. JETP **39**, 565 (1974)].

[15] Yu. M. Izyumov and V. M. Medvedev, Teoriya magnitouporyadochennykh kristallov s primesyami (Theory of Magnetically Ordered Crystals Containing Impurities). M.: Nauka, 1970, Chap. I. (Transl., Plenum Pub., 1973).

[16] M. A. Ivanov, Yu. G. Pogorelov, and M. I. Botvinko, Zh. Eksp. Teor. Fiz. **70**, 610 (1976) [Sov. Phys. JETP **43**, 317 (1976)].

[17] P. W. Anderson, Phys. Rev. **109**, 1492 (1958).

[18] M. A. Ivanov, Yu. G. Pogorelov, V. M. Loktev, K. N. Kocharyan, A. S. Prokhorov, and E. G. Rudashevsky, Solid State Commun. **33**, 623 (1980).

[19] V. M. Naumenko, V. V. Eremenko, V. M. Bandura, and V. V. Pishko, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 436 (1980) [JETP Lett. **32**, 412 (1980)].

[20] S. J. Allen, Jr. and H. J. Guggenheim, Phys. Rev. B **4**, 937 (1971).

[21] P. Martel, R. A. Cowley, and R. W. H. Stevenson, Can. J. Phys. **46**, 1355 (1968).

Translated by W. F. Brown, Jr.