Dependence of the resonant frequencies of antiferromagnets on the magnetic field, and antiferromagnetic resonance in CoF_2

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We investigated AFMR in CoF_2 in the range 0.3–4.3 mm in a field up to 250 kOe directed along the C_4 axis, at $T=4.2$°K. Experiment revealed a nonlinearity in the $\omega=\omega\ (H)$ dependence; this nonlinearity is attributed to a transverse Dzyaloshinskiĭ interaction (DI) in the form $M_xL_y+M_yL_x$ ($\mathbf{M}=\mathbf{M}_1+\mathbf{M}_2$, $L=\mathbf{M}_1-\mathbf{M}_2$). As $\omega\to 0$, the derivative $\partial\omega/\partial H$ tends to a finite value $(4.3\pm0.2)\times10^{10}$ (kOe-sec)$^{-1}$, in contradiction to spin-wave theory, which predicts $\partial\omega/\partial H\ (\omega\to 0)\to\infty$. A theoretical analysis shows that a finite value of $\partial\omega/\partial H\ (\omega\to 0)$ follows in principle from the connection between the transverse DI and the longitudinal DI $[(\mathbf{L}\cdot\mathbf{M})L_xL_y$ and $(\mathbf{L}\cdot\mathbf{M})M_xM_y]$ at $\chi_{zz}(T\to0\,°\mathrm{K})\neq0$, as is observed in experiment. The connection between the longitudinal DI and the transverse DI reflects the fact that the term of fourth degree in the magnetization in the thermodynamic potential follow from a bilinear spin Hamiltonian that determines also the transverse DI, and the contribution of the mean values of the products of the components of the fourth-degree spin operator is negligibly small, as should be the case for the spin 3/2 of divalent cobalt. The theoretical analysis is based on a total potential with allowance for all the possible symmetries of the invariants, without the use of a series expansion. As a result of the analysis, a potential of simpler form is proposed, describing fully the experiment and containing only six phenomenological parameters. Addition of arbitrary symmetry-allowed invariants to the proposed potential changes neither the form of the spectrum nor the number of the experimentally determined parameters.

Antiferromagnetic cobalt fluoride is one of the most interesting objects for the investigation of the influence of spin-orbit interactions on antiferromagnetic properties. The incompletely quenched orbit of the magnetic ion in CoF_2 is combined with a relatively large spin, equal to 3/2. As a result, a nonzero parallel susceptibility is observed in CoF_2 even at the lowest temperatures, and the interactions that are biquadratic in the spin components are of the same order as the bilinear interactions. Therefore the use of the well-developed theory of spin waves for the interpretation of the experimental results on CoF_2 becomes doubtful. The nonzero parallel susceptibility exerts an appreciable influence, e.g., on the high-frequency properties of antiferromagnets.

Antiferromagnetic resonance (AFMR) was first observed in CoF_2 by Richards [1] in a stationary magnetic field up to 50 kOe in the wavelength range 0.2–0.3 mm. When an external magnetic field is applied along the [001] axis of the crystal the AFMR frequency exhibits a magnetic-field dependence typical of easy-axis antiferromagnets. It was believed that this linear dependence is preserved up to sublattice-flipping fields. Our investigations of AFMR in CoF_2 at fields and frequencies starting from the lowest ones and ending with fields exceeding the flipping field have revealed a number of significant hitherto unknown singularities. The AFMR in CoF_2 was investigated at $T = 4.2$°K in magnetic fields up to 250 kOe and in the wavelength interval from 300 μ to 4.3 mm. Backward-wave generators were used [2]. A flow-through type microwave spectrometer, operating in a stationary magnetic field up to 150 kOe, was developed for the measurements in the range from 300 μ to 1 mm. Measurements in fields stronger than 150 kOe were performed in pulsed solenoids [3]. For the investigations in the pulsed magnetic fields, a spectrometer of the reflex type was developed for the 1–4.3 mm band, using dielectric quartz waveguides to exclude eddy currents [4]. The CoF_2 samples in the form of plates measuring 1.5×2.5×0.8 mm were glued to the end of a quartz waveguide placed at the center of the pulsed solenoid. The magnetic field was measured by precision integration (with accuracy not worse than 0.3%) of a signal from a measuring coil placed in the same plane as the investigated sample, the signal being calibrated by electron paramagnetic resonance (EPR) in DPPH located alongside the sample. This method of measuring the magnetic field determined the position of the absorption lines in the magnetic field with accuracy not worse than 2%.

The magnetic field was directed along [001] (the z axis). The measurement results are shown in Fig. 1. On the short-wave side, our results agree well, within the limits of experimental error, with the results of Richards [1,5]. In strong magnetic fields, however, a nonlinearity was observed in the dependence of ω on H. Special attention should be called to the slope of the curve near zero frequency, where the derivative $\partial\omega/\partial H(H\to H_c)$ is finite and we have $\partial\omega/\partial H(H=H_c)=(4.3\pm0.2)\times10^{10}$ (kOe-sec)$^{-1}$ (this does not agree with spin-wave theory).

The use of the simplest model potential for antiferromagnetic CoF_2 (space group D_{4h}^{14}) with a Dzyaloshinskiĭ interaction [6,7], which takes into account only interactions bilinear in the spins of the atoms

$$\Phi=\tfrac{1}{2}BM^2+\tfrac{1}{2}aL_z^2+e(M_xL_y+M_yL_x)+\tfrac{1}{2}bM_z^2-\mathbf{MH} \qquad (1)$$

(in the spin-wave approximation $M^2+L^2=\mathrm{const}$ $(\mathbf{M}\cdot\mathbf{L})=0$, $\mathbf{M}=\mathbf{M}_1+\mathbf{M}_2$, $\mathbf{L}=\mathbf{M}_1-\mathbf{M}_2$, where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations) actually leads to the nonlinear relation [5]

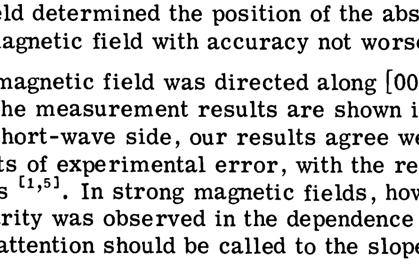

FIG. 1. Dependence of the low-frequency AFMR branch on the magnetic field in CoF_2 at T = 4.2° K.

$$\omega^2=\gamma^2\{[(H_aH_E)^{1/2}\pm H]^2-H_D^2\}, \qquad (2)$$

where the anisotropy field is $H_a=-a$, the exchange field is $H_E=B+b$, and the Dzyaloshinskiĭ field is $H_D=e$ (we note that in the model that takes into account the expansion terms up to second order in the spins there are six constants: $(M_1^0)^2$, $(M_2^0)^2$, B, a, e, and b, and these must be determined experimentally).

However, this very simple model, which describes well the high-frequency properties of other antiferromagnets [8-10] (e.g., MnF_2), does not describe even qualitatively the results of AFMR experiments in CoF_2. Thus, e.g., it follows from the theoretical model (1) that $\partial\omega/\partial H(\omega\to 0)=\infty$, which contradicts aforementioned experimental results (Fig. 1).

This raises the problem of finding a theoretical model that describes adequately the experimental results on cobalt fluoride. To construct such a theory we shall use, as is customary, the Landau theory of phase transitions [11].

The thermodynamic potential of the Landau theory, as a function of the vectors **M** and **L**, is invariant with respect to the symmetry group D_{4h}^{14} of the paramagnetic phase, and can consequently depend only on the following thirteen invariant combinations of the components of the vectors **M** and **L** [12]:

$$\begin{gathered} I_1=\mathbf{L}^2,\ I_2=\mathbf{M}^2,\ I_3=(\mathbf{LM})^2,\ I_4=L_z^2,\ I_5=M_z^2,\\ I_6=(\mathbf{LM})L_zM_z,\ I_7=L_xM_y+L_yM_x,\ I_8=(\mathbf{LM})L_xL_y,\\ I_9=L_xL_yL_zM_z,\ I_{10}=L_x^2L_y^2,\ I_{11}=(\mathbf{LM})M_xM_y,\\ I_{12}=L_zM_zM_xM_y,\ I_{13}=M_x^2M_y^2. \end{gathered} \qquad (3)$$

The dependence of Φ on the invariants I_1, I_2, and I_3, as seen from their symmetry, is determined by the exchange interactions; the dependence on the invariants I_4, I_5, and I_6 is connected with relativistic interactions that determine the uniaxial anisotropy in CoF_2. The remaining invariants are peculiar to the given magnetic structure, and the dependence on them is due to relativistic interactions that determine the anisotropy in the basal plane (the plane perpendicular to C_4). In particular, the dependence on I_7 is determined by the bilinear Dzyaloshinskiĭ interaction [6,7]. The invariants I_8 and I_{11} describe the longitudinal weak Dzyaloshinskiĭ ferromagnetism [6,13], I_9 and I_{10} describe the anisotropic increment to I_8 and I_{11}, while the invariants I_{10} and I_{13} correspond to anisotropy in the basal plane.

Using these thirteen invariants, we can write down a series expansion of the thermodynamic potential up to any power in the spin components. For example, accurate to terms of second order, the thermodynamic potential takes the form [6,7]

$$\Phi={}^1/_2AI_1+{}^1/_2BI_2+{}^1/_2aI_4+{}^1/_2bI_5+eI_7. \qquad (1a)$$

The series containing fourth-order terms in the spin components, however, contains already 38 terms and to work with such an expansion is technically difficult. In the general treatment we shall therefore simply regard the thermodynamic potential as a function of the invariants, $\Phi=\Phi(I_1, I_2, \ldots, I_{13})$, and the expansion will be spelled out specifically only after separating those interactions that must be taken into account in the model in order to describe the experiment fully.

Thus, the thermodynamic potential in a magnetic field takes the form

$$\tilde{\Phi}=\Phi(I_1, I_2, \ldots, I_{13})-\mathbf{MH}. \qquad (1b)$$

From this we find that in the state $\mathbf{M}\parallel\mathbf{L}\parallel\mathbf{H}\parallel C_4$ the values of M and L are determined by a system of two nonlinear equations

$$\begin{gathered} 2M[\Phi_2+\Phi_5+M^2(\Phi_3+\Phi_6)]=H,\\ 2L[\Phi_1+\Phi_4+M^2(\Phi_3+\Phi_6)]=0, \end{gathered} \qquad (4)$$

where $\Phi_k=\partial\Phi/\partial I_k$.

From this we can obtain the dependence of M and L on the magnetic field (for example, accurate to H^3):

$$\begin{gathered} M=\chi_0H\left\{1-4\chi_0^3H^3\left[B_2^0+L_0^2C_1^0+\frac{A_1^0+\alpha_0}{B_1^0}(A_1^0+\alpha_0+L_0^2C_1^0)\right]\right\},\\ L=L_0-\chi_0^2H^2\frac{A_1^0+\alpha_0}{2L_0B_1^0}, \end{gathered} \qquad (5)$$

where we have introduced the notation

$$\begin{gathered} \alpha_0=\Phi_3^0+\Phi_6^0,\ A_1^0=2(\Phi_{12}^0+\Phi_{15}^0+\Phi_{42}^0+\Phi_{45}^0),\\ B_1^0=\Phi_{11}^0+2\Phi_{14}^0+\Phi_{44}^0,\ B_2^0=\Phi_{22}^0+2\Phi_{52}^0+\Phi_{55}^0,\\ C_1^0=\Phi_{32}^0+\Phi_{35}^0+\Phi_{62}^0+\Phi_{65}^0;\\ \chi_0=[2(\Phi_2^0+\Phi_5^0+\alpha_0L_0^2]^{-1},\ \Phi_{ik}=\partial^2\Phi/\partial I_i\partial I_k. \end{gathered}$$

The index zero (e.g., A_1^0, B_1^0, L_0, χ_0, etc.) signifies that this parameter (constant) is taken at $H=0$. The field dependence of the phenomenological parameters $R=\{A_1, B_1, B_2, C_1, \alpha\}$ is determined by the formula

$$R=R^0+\chi_0^2H^2\left[R_2^0+R_5^0-\frac{A_1^0+\alpha_0}{B_1^0}(R_1^0+R_4^0)\right],$$

where $R_i^0=\partial R/\partial I_i$ at $H=0$.

The symbols α, A_1, B_1, B_2, C_1, and χ were introduced because none of the results of the theory contain the phenomenological parameters Φ_i and Φ_{ik} separately, but only in the combinations given above. This makes it possible to determine which constants should be retained in the model expansion of the thermodynamic potential in order to describe a particular effect without loss of generality. Thus, if a certain effect is proportional to C_1, then to describe this effect it is necessary to take into account in the model expansion of the thermodynamic potential the terms of not less than the sixth power in the spin components, and it suffices to retain in expansion of Φ in powers of the spins any of the four invariants I_6I_5, I_3I_2, I_3I_5, and I_6I_2.

To describe the dynamic properties of magnets, it is important to make the right choice of the equations of motion. As a rule, the Landau-Lifshitz equations are used [14], and for antiferromagnets they take the form

$$\begin{gathered} \frac{1}{\gamma}\dot{\mathbf{M}}=\left[\mathbf{M}\times\frac{\partial\Phi}{\partial\mathbf{M}}\right]+\left[\mathbf{L}\times\frac{\partial\Phi}{\partial\mathbf{L}}\right],\\ \frac{1}{\gamma}\dot{\mathbf{L}}=\left[\mathbf{M}\times\frac{\partial\Phi}{\partial\mathbf{L}}\right]+\left[\mathbf{L}\times\frac{\partial\Phi}{\partial\mathbf{M}}\right]. \end{gathered} \qquad (6)$$

For small deviations from the equilibrium position, the equations can be linearized and represented in the form $d\mathbf{X}/dt=\hat{\gamma}\mathbf{Y}$ or, written out fully,

$$\frac{d}{dt}\begin{pmatrix} m_x\\ m_y\\ m_z\\ l_x\\ l_y\\ l_z \end{pmatrix}=\begin{pmatrix} 0 & gM_z^0 & -gM_y^0 & 0 & gL_z^0 & -gL_y^0\\ -gM_z^0 & 0 & gM_x^0 & -gL_z^0 & 0 & gL_x^0\\ gM_y^0 & -gM_x^0 & 0 & gL_y^0 & -gL_x^0 & 0\\ 0 & gL_z^0 & -gL_y^0 & 0 & gM_z^0 & -gM_y^0\\ -gL_z^0 & 0 & gL_x^0 & -gM_z^0 & 0 & gM_x^0\\ gL_y^0 & -gL_x^0 & 0 & gM_y^0 & -gM_x^0 & 0 \end{pmatrix}\begin{pmatrix} E_x\\ F_y\\ F_z\\ N_x\\ N_y\\ N_z \end{pmatrix}, \qquad (7)$$

where $\mathbf{X}=\{m_x, \ldots, l_z\}$ are small deviations from the equilibrium components of the vectors **M** and **L**;

$\mathbf{Y} = \{F_X, \ldots, N_Z\}$ are the thermodynamic forces $\mathbf{Y} = \hat{\alpha}\mathbf{X}$ that result from the corresponding deviations from the equilibrium position [11]:

$$\begin{pmatrix} F_x \\ F_y \\ F_z \\ N_x \\ N_y \\ N_z \end{pmatrix} = \begin{pmatrix} \partial^2\Phi/\partial M_x\partial M_x \ldots & \ldots \partial^2\Phi/\partial M_x\partial L_z \\ \cdot \quad \ldots & \cdot \\ \cdot \quad \ldots & \cdot \\ \cdot \quad \ldots & \cdot \\ \cdot \quad \ldots & \cdot \\ \partial^2\Phi/\partial L_z\partial M_x \ldots & \ldots \partial^2\Phi/\partial L_z\partial L_z \end{pmatrix} \begin{pmatrix} m_x \\ m_y \\ m_z \\ l_x \\ l_y \\ l_z \end{pmatrix}. \tag{8}$$

All second derivatives in the matrix $\hat{\alpha}$ are taken in the equilibrium state.

The region where the matrix $\hat{\alpha}$ is positive-definite as a function of H and T determines completely the stability of the equilibrium state of the magnetic subsystem. Thus, the motion is described by a product of two matrices $\hat{\gamma}\hat{\alpha}$, one of which is connected with the form of the equations of motion and the other with the equilibrium condition. However, as noted by the authors of [14], relations (6) are invariant against arbitrary rotations of the spin system as a unit, and therefore correspond only to the exchange approximation. In this approach, the anisotropic interactions can be accounted for only via the stability matrix $\hat{\alpha}$. It is known, however, that to explain the experiment in many cases it is necessary to take into account the anisotropy also in the matrix $\hat{\gamma}$, e.g. with the aid of the anisotropic g-factor [8,15].

The cobalt fluoride investigated by us is different because the exchange interactions are comparable with the anisotropic interactions, so that for a complete analysis of the dynamic properties it is expedient to take rigorous account of the anisotropy in $\hat{\gamma}$ in most complete fashion. Allowance for the anisotropy of $\hat{\gamma}$ by introducing an anisotropic g-factor is not the most complete way. Great promise is offered in this case by nonequilibrium thermodynamics [16]. Indeed, if we write down the symmetry-allowed Onsager equations for the magnetic subsystem of cobalt fluoride, then the antisymmetrical matrix $\hat{\gamma}$ takes the form

$$\begin{vmatrix} 0 & \gamma_1 M_z^0 & \gamma_2 M_y^0 + \gamma_3 L_x^0 & \lambda_1 M_z^0 & \lambda_2 L_z^0 & \lambda_3 M_x^0 + \lambda_4 L_y^0 \\ & 0 & -(\gamma_2 M_x^0 + \gamma_3 L_y^0) & -\lambda_2 L_z^0 & -\lambda_1 M_z^0 & -(\lambda_3 M_y^0 + \lambda_4 L_x^0) \\ & & 0 & -\lambda_5 M_x^0 + \lambda_6 L_y^0 & \lambda_5 M_y^0 - \lambda_6 L_x^0 & 0 \\ & & & 0 & -\rho_1 M_z^0 & -(\rho_2 M_y^0 + \rho_3 L_x^0 \\ & & & & 0 & \rho_2 M_x^0 + \rho_3 L_y^0 \\ & & & & & 0 \end{vmatrix}. \tag{9}$$

This matrix goes over into the matrix $\hat{\gamma}$ for equations that take into account the anisotropy only via the anisotropic g factor, if we put

$$\gamma_1 = -\lambda_2 = -\rho_1 = \tilde{g}_\perp,\ \lambda_1 = 0,\ \gamma_2 = \lambda_4 = -\lambda_6 = -\rho_2 = -\tilde{g}_\parallel,$$
$$\gamma_3 = \lambda_3 = \lambda_5 = -\rho_3 = 2\tilde{g}_\parallel \tilde{\tau}.$$

The agreement between $\tilde{g}_\perp$, $\tilde{g}_\parallel$, and $\tilde{\tau}$, and the notation introduced by Turov [8] is determined by the relations

$$\tilde{g}_\parallel = g_\parallel \frac{1+\tau^2}{1-\tau^2}, \quad \tilde{\tau} = \frac{\tau}{1+\tau^2}, \quad \tilde{g}_\perp = \frac{g_\perp^2}{g_\parallel}(1-\tau^2), \quad \tau = \frac{g_{xy}}{g_\perp}.$$

The Landau-Lifshitz equations in the linear approximation can be obtained by putting $\tilde{g}_\perp = \tilde{g}_\parallel = \tilde{g}$ and $\tilde{\tau} = 0$. Regardless of which equations of motion are used to determine the resonant frequencies, the following relation holds:

$$\omega^6 - K_2\omega^4 + K_4\omega^2 - K_6 = 0, \qquad K_6 = \det\hat{\nu} = \det\hat{\gamma}\hat{\alpha} = \det\hat{\gamma}\det\hat{\alpha}, \tag{10}$$

where K_4 is the sum of all the possible determinants of the fourth-rank matrices, obtained from $\hat{\nu}$ by crossing out two lines and two columns that intersect on the principal diagonal; K_2 is the analogous sum of all the possible determinants of the second-rank matrices, obtained by crossing out from $\hat{\nu}$ four columns and four lines that intersect on the principal diagonal.

In the investigated case $\mathbf{M} \parallel \mathbf{H} \parallel \mathbf{L} \parallel z$ the rows and columns corresponding to M_Z and L_Z in the matrix $\hat{\nu}$ consist of zeros only, and Eq. (10) for the resonant frequencies takes the form

$$\omega^2[\omega^4 - K_2\omega^2 + \delta_1\delta_2] = 0, \tag{10a}$$

where $\delta_1 = \det\tilde{\tilde{\gamma}}$, $\delta_2 = \det\tilde{\tilde{\alpha}}$, and the matrices $\tilde{\tilde{\gamma}}$ and $\tilde{\tilde{\alpha}}$ are obtained from the matrices $\hat{\gamma}$ and $\hat{\alpha}$ by crossing out the rows and columns corresponding to M_Z and L_Z. We are interested in the behavior of the solution ω_1 of Eq. (10a)

$$\omega_1^2 = K_2/2 - (K_2^2/4 - \delta_1\delta_2)^{1/2}, \tag{11}$$

which corresponds to the experimental curve given in Fig. 1. The vanishing of ω_1, as seen from (11), can be due to the vanishing of either δ_1 or δ_2. In the former case, as $\delta_1 \to 0$, we see that

$$\left.\frac{\partial\omega_1}{\partial H}\right|_{\omega_1\to 0} \approx \left(\frac{\delta_2}{\delta_1}K_2\right)^{1/2}\frac{\partial\delta_1}{\partial H}, \tag{12}$$

$$\delta_1 = [(\gamma_1\rho_1 - \lambda_1^2)M^2 - \lambda_2^2 L^2]^2. \tag{13}$$

Hence

$$\frac{\partial\delta_1}{\partial H} = 4\delta_1^{1/2}\left[(\gamma_1\rho_1 - \lambda_1^2)M\frac{\partial M}{\partial H} - \lambda_2^2 L\frac{\partial L}{\partial H}\right]. \tag{14}$$

The quantity in the square brackets in (14) is finite and consequently, if the vanishing of the frequency $\omega_1(H)$ is due to the vanishing of δ_1, then $\partial\omega_1/\partial H(\omega_1 \to 0)$ is a finite quantity. If we use the Landau-Lifshitz equations with an anisotropic g-factor, then

$$\delta_1 = \tilde{g}_\perp^4(L_z^2 - M_z^2)^2. \tag{13a}$$

Therefore, if the frequency tends to zero like $\delta_1^{1/2}$, then M_Z in the field in which $\omega_1 = 0$ should coincide with L_Z. It appears that this does not agree with the results of measurements of magnetization in a parallel field [17]. It is clear therefore that in a model in which the fact that $\partial\omega_1/\partial H(\omega_1 \to 0) \neq \infty$ is attributed to the vanishing of ω_1 due to the vanishing of δ_1 (i.e., not to the loss of the stability of the state of the subsystem), the equations describing the motion of the magnetic subsystem of CoF_2 should differ significantly from the Landau-Lifshitz equations.

If ω_1 vanishes as a result of δ_1, then the function $\omega_1 = \omega_1(H)$ near $\omega_1 = 0$ should take the form shown in Fig. 2, where

$$\left.\frac{\partial\omega_1}{\partial H}\right|_{H\to H_1+0} = \left.\frac{\partial\omega_1}{\partial H}\right|_{H\to H_1-0} \neq \infty,$$

and the field H_2 is determined by the conditions for the stability of the state $\mathbf{M} \parallel \mathbf{L} \parallel \mathbf{H} \parallel C_4$ (i.e., by the condition that the matrix α be positive-definite). Since this fact

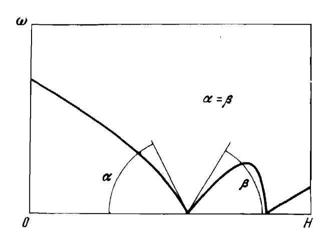

FIG. 2. Dependence of the AFMR frequency on the magnetic field. The vanishing of the frequency is connected with the matrix $\|\gamma_{ik}\|$ of the equations of motion.

is not observed experimentally, let us consider a different possibility of the vanishing of ω_1, connected with the vanishing of δ_2. In this case we have at $\mathbf{L} \parallel \mathbf{M} \parallel \mathbf{H} \parallel z$

$$\left.\frac{\partial\omega_1}{\partial H}\right|_{\omega_1\to 0}=\left(\frac{\delta_1}{\delta_2}K_2\right)^{1/2}\frac{\partial\delta_2}{\partial H}; \tag{15}$$

$$\begin{aligned}\delta_2=&\{4\Phi_1\Phi_2-\Phi_7^2-L^2M^2[(2\Phi_3+\Phi_6)^2-(\Phi_8+\Phi_9)(\Phi_{11}+\Phi_{12})]\}^2\\&-4L^2M^2[\Phi_1(\Phi_{11}+\Phi_{12})+\Phi_2(\Phi_8+\Phi_9)-\Phi_7(2\Phi_3+\Phi_6)]^2=T_1^2-T_2^2=UV,\end{aligned} \tag{16}$$

where U and V are the sum and difference of the square roots of the first and second terms. Let the vanishing of δ_2 be connected with the vanishing of U. Then

$$\left.\frac{\partial\omega_1}{\partial H}\right|_{\omega_1\to 0}\approx\left(\frac{\delta_1 VK_2}{U}\right)^{1/2}\frac{\partial U}{\partial H}$$

and in order for $\partial\omega_1/\partial H$ near $\omega_1=0$ not go off to infinity, it is necessary to have $\partial U/\partial H(\omega_1\to 0)\sim U^{1/2}$, i.e., at the very point where the state $\mathbf{L}\parallel\mathbf{M}\parallel\mathbf{H}\parallel C_4$ loses stability we should have

$$U=0, \tag{17}$$

$$\partial U/\partial H=0. \tag{18}$$

We assume, in accordance with the available experimental data [17], that $M \ll L$ near the stability loss, and write down accordingly Eqs. (17) and (18) accurate to H^2:

$$\left(\frac{\omega}{\gamma}\right)^2_{H=0}-H_c^2(\chi_{zz}^{z2}L^4E_1)_{H=H_c}=2H_c(\chi_{zz}^zL^2E_2)_{H=H_c}, \tag{17a}$$

$$\left(\frac{1}{\gamma^2}\frac{\partial\omega^2}{\partial H}\right)_{H=0}-2H_c(\chi_{zz}^{z2}L^4E_1)_{H=H_c}=2(\chi_{zz}^zL^2E_2)_{H=H_c}, \tag{18a}$$

$$\begin{aligned}L^4E_1&=(1/\chi_{zz}^z-1/\chi_{xx}^y)^2+\frac{1}{L^2}H_{d\parallel}^{(1)}H_{d\parallel}^{(2)},\\L^3E_2&=H_AH_{d\parallel}^{(2)}+H_EH_{d\parallel}^{(1)}+H_{d\perp}L(1/\chi_{zz}^z-1/\chi_{xx}^y).\end{aligned} \tag{19}$$

Here and below the symbol χ_{ii}^k denotes the susceptibility along the i axis, when the component of the vector $\mathbf{L}$ is directed along the k axis;

$$2\Phi_1S=-2\Phi_3S=H_A,\quad S=(L^2+M^2)^{1/2}\neq\text{const},\quad 2\Phi_2=\chi_{xx}^y,$$

$\Phi_7S\equiv H_{d\perp}$ is the perpendicular Dzyaloshinskiĭ field in the cobalt fluoride, measured at $\mathbf{L}\parallel y$, $\mathbf{H}\parallel x$; $\Phi_3 \equiv 1/\chi_{zz}^z-1/\chi_{xx}^y$. In addition, we have separated two longitudinal Dzyaloshinskiĭ fields at $\mathbf{L}\parallel z$:

$$H_{d\parallel}^{(1)}=(\Phi_8+\Phi_9)L^3,$$

$$H_{d\parallel}^{(2)}=(\Phi_{11}+\Phi_{12})L^3$$

and take account of the fact that, accurate to H^2,

$$\left.\frac{\partial(\Phi_1\Phi_2-\Phi_7^2)}{\partial H}\right|_{H=H_c}=\left.\frac{\partial\omega_1^2}{\partial H}\right|_{H=0}$$

We see that Eqs. (17a), (18a), and (19) yield a method of determining two effective fields in CoF_2, namely $H_{d\parallel}^{(1)}$ and $H_{d\parallel}^{(2)}$, which cannot be determined from other experiments. The existence of the relations (17a), (18), and (19) between $\Phi_8+\Phi_9$, $\Phi_{11}+\Phi_{12}$, and the other properties of the theory, determines the fact that $\partial\omega_1/\partial H(\omega_1=0)\neq\infty$ in a field that violates the condition for the stability of the state $\mathbf{L}\parallel\mathbf{M}\parallel C_4$. It is clear from the form of I_8 and I_9 or I_{11} and I_{12} that the dependence on these quantities (i.e., on $\Phi_8+\Phi_9$ or $\Phi_{11}+\Phi_{12}$) is determined by the longitudinal weak Dzyaloshinskiĭ ferromagnetism. It follows from this unambiguously that if the vanishing of the frequency ω_1 in CoF_2 is due to the loss of stability of the state $\mathbf{L}\parallel\mathbf{M}\parallel C_4$, then the fact that $\partial\omega_1/\partial H(\omega_1=0)\neq\infty$ can be attributed only to the simultaneous presence of a nonzero parallel susceptibility $(\mathbf{L}\cdot\mathbf{M}\neq 0)$ and a longitudinal Dzyaloshinskiĭ interaction $[(\mathbf{LM})L_xL_y\neq 0,\ (\mathbf{LM})M_xM_y\neq 0]$ in the magnetic subsystem of CoF_2. If this explanation

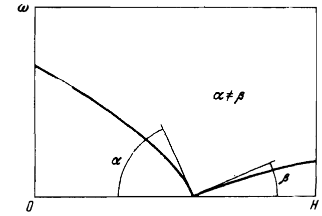

FIG. 3. Dependence of the AFMR frequency on the magnetic field. The vanishing of the frequency is connected with the stability matrix $\|\alpha_{ik}\|$.

is accepted, then $\omega_1=\omega_1(H)$ near $\omega_1=0$ takes the form shown in Fig. 3, with $\partial\omega_1/\partial H(H\to H_c+0)\neq\partial\omega_1/\partial H(H\to H_c-0)$.

A comparison of the theoretical analysis with the experimental results indicates that the loss of the stability of the state $\bar{\mathbf{L}}\parallel\bar{\mathbf{M}}\parallel C_4$ is a more probable cause of the vanishing of the frequency in CoF_2 than is the vanishing of δ_1. This explanation of the fact that $\partial\omega_1/\partial H(\omega_1\to 0)\neq\infty$ may seem to be artificially related to the stability of the state $\mathbf{L}\parallel\mathbf{M}\parallel C_4$. Actually (17a), (18a), and (19) require a certain connection between the parameters of the theory, a connection that does not follow from the thermodynamic relations. Without dwelling on the microscopic premises of such a connection, we shall show that the fact $\partial\omega_1/\partial H(\omega_1=0)\neq\infty$ cannot take place if no definite relations that follow from the thermodynamic equations exist between the parameters of the theory. Assume that at ω_1 close to zero the state $\mathbf{L}\parallel\mathbf{M}\parallel C_4$ goes over into a state in which the vectors $\mathbf{M}=(M_x, M_y, M_z)$ and $\mathbf{L}=(L_x, L_y, L_z)$ have all the components different from zero. This variant is admitted when considering the transitions that are possible in a magnetic field, for $\tilde{\Phi}=\Phi(I_1, I_2, \ldots, I_{13})-\mathbf{MH}$, if $L^2+M^2\neq\text{const}$, $(\mathbf{LM})\neq\text{const}$. We shall show that in this case, too, the result $\partial\omega_1/\partial H(\omega_1\to 0)\neq\infty$ calls for definite relations between the phenomenological parameters of the theory.

If we consider the general symmetry-allowed equations of motion, then this statement is obvious, since [16]

$$2\omega_1\frac{\partial\omega_1}{\partial H}(\omega_1=0)=\frac{\partial K_6}{\partial H}=\frac{\partial|\gamma_{ik}||\alpha_{ik}|}{\partial H},\quad|\gamma_{ik}|=G^2,$$

and all the arguments presented above can be repeated for two variants: 1) $|\gamma_{ik}|=0$ (in analogy with δ_1) and 2) $|\alpha_{ik}|=0$ (in analogy with δ_2). We then obtain in the former case the previously unobserved relation $\omega=\omega(H)$ (Fig. 2), and in the second variant we obtain between the parameters of the theory relations that are not required by symmetry. On the other hand, if we assume that the motion of the magnetic moments of the sublattices obeys the Landau-Lifshitz equations with anisotropic g-factor, then for an analysis of the lower branch of the spectrum it is necessary to analyze the relations $K_4=0$ and $\partial K_4/\partial H(K_4\to 0)=0$. The analysis has shown that in this case two relations, which do not follow from thermodynamics, are obtained between the parameters of the potential and the phenomenological values of the components of the tensor g_{ik}. Therefore the simplest theory that explains the fact that $\partial\omega_1/\partial H(\omega_1\to 0)\neq\infty$, without assuming an unusual (Fig. 2) form of the dependence of the frequency on the field, should of necessity take into account the longitudinal weak Dzyaloshinskiĭ ferromagnetism.

Thus, this possibility of vanishing of the frequency with $\partial\omega_1/\partial H(\omega_1\to 0)\neq\infty$ seems to us to agree better with experiment. An analysis that starts out from the most general form of the potential Φ makes it possible to write for $\mathbf{H} \parallel Z$ a very simple form of the potential, which describes fully the experiment on CoF_2 and contains only six experimentally-determined parameters (taking into account the relations (17a) and (18a)):

$$\Phi = {}^1/_2 AL^2 + {}^1/_2 BM^2 + {}^1/_2 D_1 (\mathbf{LM})^2 + {}^1/_2 aL_z^2 + {}^1/_4 CL^4$$
$$+ d_\perp (L_x M_y + L_y M_x) + d_\parallel^{(1)} (\mathbf{LM}) L_x L_y + d_\parallel^{(2)} (\mathbf{LM}) M_x M_y - M_z H_z,$$

i.e., as many parameters as are used in the model description with the bilinear potential. The presented potential is complete in the sense that addition of any other symmetry-allowed invariants does not change either the form of the $\omega = \omega(H)$ dependence or the number of the experimentally determined parameters.

The foregoing relations between $H_{d\perp}$, $H_{d\parallel}^{(1)}$, and $H_{d\parallel}^{(2)}$ can be interpreted in the following manner. A fraction of the energy describing the longitudinal Dzyaloshinskiĭ interaction is expressed in terms of the mean values of the components of the spin operators and is derived from the spin Hamiltonian in two ways. First, this part of the energy appears as the mean value of the spin Hamiltonian of fourth power in the components of the spin operators and, second, it is derived in third order perturbation theory from the bilinear spin Hamiltonian with allowance for the intra-atomic spin-orbit interaction. The obtained relations between $H_{d\perp}$, $H_{d\parallel}^{(1)}$, and $H_{d\parallel}^{(2)}$ show that the average products of the fourth-degree spin-operator components are small in comparison with the analogous terms obtained from the bilinear Hamiltonian in third order perturbation theory, as should be the case also for spin 3/2. Thus, the experimentally observed connection between the longitudinal and transverse Dzyaloshinskiĭ interaction, which becomes manifest in the inequality $\partial\omega/\partial H(\omega \to 0) \neq \infty$, agrees with the spin value 3/2 for divalent cobalt.

It should be noted that the experiment reported above and the observation of $\partial\omega/\partial H(\omega \to 0) \neq \infty$ can be regarded as experimental proof of the existence of longitudinal weak ferromagnetism in antiferromagnets.

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