

Anomalies of high-frequency magnetic permeability of hematite at the Morin phase transition

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The submillimeter magnetic spectra of hematite and of hematite with a Ga^{3+} impurity ($x \approx 5\text{--}6\%$) are investigated in the frequency range $2\text{--}16\text{ cm}^{-1}$ at $T = 4.2\text{--}600\text{ K}$. The temperature dependence of the resonant frequency $\nu_0(T)$, of the contribution of the mode to the static magnetic permeability $\Delta\mu(T)$, and of the linewidth $\Gamma(T)$ are obtained for the high-frequency antiferromagnetic resonance mode. Discontinuities in the plots of $\Delta\mu(T)$ and $\Gamma(T)$ are observed at the Morin point. An anomalous increase of $\Gamma(T)$ at liquid-helium temperatures is observed for the hematite containing no gallium. It is shown that the experimentally observed $\Delta\mu(T)$ and $\nu_0(T)$ can be satisfactorily explained by using a simple model based on the Landau-Lifshitz equations with a thermodynamic potential that includes the first two anisotropy constants.

I. INTRODUCTION

Hematite ($\alpha\text{-Fe}_2\text{O}_3$) is an antiferromagnet having a Néel temperature $T_N \approx 950\text{ K}$ and a rhombohedral symmetry described by the space group D_{3d}^6 . At $T_M = 263\text{ K}$ it undergoes an orientational first-order phase transition from a weakly ferromagnetic easy-plane state ($T > T_M$) to an antiferromagnetic easy-axis state ($T < T_M$) (see, e.g., Ref. 1). The temperature of the phase transition in hematite can be varied in a wide range by replacing a small fraction of the Fe^{3+} ions by diamagnetic impurity ions such as Al^{3+} , Ga^{3+} , and others.² All this, in conjunction with the relative simplicity of the crystal and magnetic structure, makes hematite a convenient model for the study of the behavior (anomalies) of various physical quantities in the course of a first-order phase transition.

The static magnetic properties of hematite, which have been investigated in detail (see, e.g., Refs. 2–7), are adequately described by a thermodynamic theory⁸ that takes the first and second anisotropy constants into account (see also Refs. 1 and 9). This same model, as shown in Ref. 10, also describes the behavior of the antiferromagnetic-resonance (AFMR) frequencies. For $T > T_M$ hematite has two AFMR modes—quasiferromagnetic (low-frequency) and quasiantiferromagnetic (high-frequency). The variation of the frequencies of these modes as functions of magnetic field and temperature has been investigated in detail in Refs. 10–19.

The purpose of the present paper is a study of the temperature dependence of the parameters of the high-frequency AFMR branch by a new experimental method that yields, besides the frequency, two other magnon-mode parameters, viz., the damping and the contribution to the static magnetic permeability. The measurements were performed in the temperature interval $4.2\text{--}660\text{ K}$ and at frequencies $\nu = 2\text{--}16\text{ cm}^{-1}$. We investigated samples of pure hematite and of hematite doped with Ga^{3+} .

2. EXPERIMENT

The measurements were made with an “Epsilon” submillimeter BWO spectrometer²⁰ by the quasi-optical proce-

dure described in Refs. 21 and 22. The working radiation was monochromatic to within $\sim 0.001\text{ cm}^{-1}$, the degree of polarization was $\sim 99.99\%$, and the excess of power above the receiver noise level was $\sim 10^5$.

In the course of the measurements the spectrometer records the transmittances and reflectances of plane-parallel samples. The end results are the spectra of the dielectric constant and of the magnetic permeability of the substance being investigated.

Single crystals of hematite and of hematite doped with $5\text{--}6\%$ gallium were grown from a solution in molten $\text{Bi}_2\text{O}_3\text{--Na}_2\text{O}$. The Morin temperature of the resulting hematite single crystals was $258 \pm 0.5\text{ K}$. The slight decrease of T_M in the hematite is apparently due to uncontrollable cobalt- and silicon-ion impurities. The samples were disks $\sim 1\text{ cm}$ in diameter and $\sim 1\text{ mm}$ thick, with planes coinciding with the basal plane of the crystal. To produce a single-domain state the sample was placed in a magnetic field of $\sim 200\text{--}400\text{ Oe}$ in the basal plane of the crystal. To excite the high-frequency (antiferromagnetic) AFMR mode the magnetic field of the wave was oriented along the constant field H_0 . Measurements have shown that the resonant frequency of the mode remains constant to within 0.04 GHz when the orientation of H_0 is varied. All the results that follow are therefore referred to a single field orientation.

Figure 1 shows typical transmission spectra $T(\nu)$ of

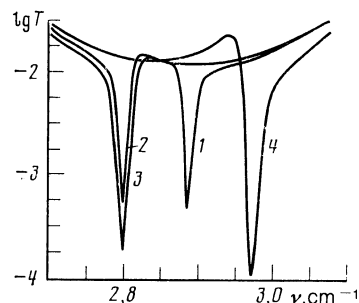


FIG. 1. Transmission spectra of hematite near the Morin transition: 1 — $T = 259\text{ K}$, 2 — 258.1 , 3 — 257.9 , 4 — 256 .

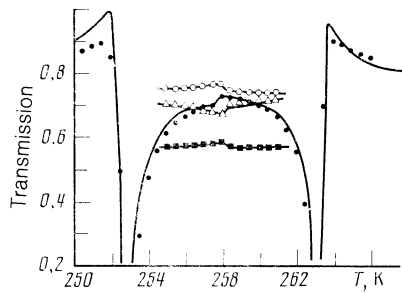


FIG. 2. Temperature dependence of the transmission coefficient of a hematite sample 1.232 mm thick near T_M at fixed frequencies: ● — $\nu = 3.23$ cm^{-1} , ○ — 2.4 cm^{-1} , △ — 2.53 cm^{-1} , ■ — 2.37 cm^{-1} ; solid lines—experiment, points—calculation.

hematite at several temperatures near the Morin transition. The observed absorption line is in fact the high-frequency AFMR mode. It can be seen that when the temperature decreases the mode shifts towards the lower frequencies, and after the transition its resonant frequency rises again. An interesting feature is the jumplike change of the sample transmittance at $T = T_M$, observed both within and outside the absorption line (curves 2 and 3 of Fig. 1). Similar “bursts” of absorption of submillimeter radiation in hematite in the Morin point were also observed earlier,¹⁹ but their nature remained unclear. For a more detailed study of this effect we measured the temperature dependences of the relative change of the transmittance at several fixed frequencies. As seen from the experimental data shown in Fig. 2, the character of the curves depends strongly on the frequency of the working radiation. Thus, at $\nu = 3.23$ cm^{-1} (●) two resonance-absorption lines and a jump in the transmittance are observed at Morin point, while at lower frequencies (○, △, ■) there are no resonance lines but the jump at $T = T_M$ remains. We point out that the jumps on the ○ and ■ curves are opposite in sign to the jumps on ● and △ curves. A more detailed analysis of these results will be presented below.

To determine the parameters of the AFMR mode, the transmission spectra of the samples were reduced by a procedure²² in which the frequency dispersion of the complex magnetic permeability is specified by the harmonic-oscillator equation

$$\mu = 1 + \Delta\mu\nu_0^2 / (\nu_0^2 - \nu^2 + i\nu\Gamma),$$

where ν_0 is the resonance frequency, Γ the line width, and $\Delta\mu$ the mode contribution to the static magnetic permeability. The temperature dependences of the mode parameters $\nu_0(T)$, $\Delta\mu(T)$, and $\Gamma(T)$ obtained in this manner are shown in Fig. 3.

The resulting $\nu_0(T)$ dependence agrees well with the data of Ref. 19. It can be seen from Fig. 3a that $\nu_0(T)$ has a minimum ($\nu_0^{\text{min}} = 2.8$ cm^{-1}) and a kink at the transition point. Near the transition point ($|T - T_M| < 7$ K) the $\nu_0(T)$ plot is linear and has the same slope $|d\nu_0/dT| = 0.086$ $\text{cm}^{-1}/\text{deg}$ above and below T_M . At low temperatures ($T \lesssim 100$ K) the frequency $\nu_0(T)$ is practically independent of T and is equal to 6.93 ± 0.01 cm^{-1} .

The contribution $\Delta\mu$ of the mode to the static magnetic permeability has an abrupt discontinuity at the Morin point (Fig. 3b). The linewidth has an unusual behavior at low temperatures and increases, starting with $T \approx 100$ K, from

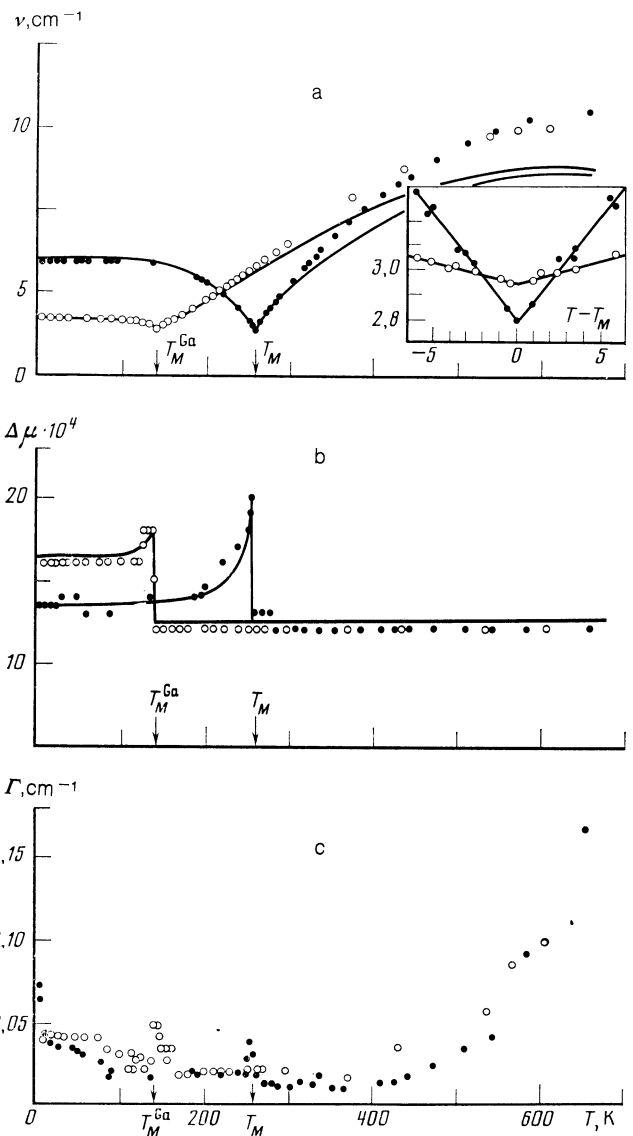


FIG. 3. a) Temperature dependence of the resonant frequencies of ● — $\alpha - \text{Fe}_2\text{O}_3$, ○ — $\alpha - \text{Fe}_{2-x}\text{Ga}_x\text{O}_3$; points—experiment, solid lines—calculation. b) Temperature dependence of the mode contributions to the static magnetic permeability: ● — $\alpha - \text{Fe}_2\text{O}_3$, ○ — $\alpha - \text{Fe}_{2-x}\text{Ga}_x\text{O}_3$; points—experiment, solid lines—calculation. c) Temperature dependences of linewidths: ● — $\alpha - \text{Fe}_2\text{O}_3$; ○ — $\alpha - \text{Fe}_{2-x}\text{Ga}_x\text{O}_3$.

0.02 to 0.07 cm^{-1} at $T = 4.2$ K (Fig. 3c). The real and imaginary parts of the dielectric constant in the investigated temperature range were $\epsilon' = 24 - 25$ and $\epsilon'' = (2 - 4) \cdot 10^{-2}$.

We consider now the behavior of the parameters of the quasiantiferromagnetic AFMR mode in hematite doped with Ga^{3+} ($T_M^{\text{Ga}} = 140$ K). It can be seen from Fig. 3 that addition of 5–6% of the diamagnetic gallium impurity changes the resonant frequency at T_M by an approximate factor of 5. The width of the resonance line in the doped hematite at $T > T_M^{\text{Ga}}$ was found to be 1.5 times larger than in pure hematite and amounted to 0.02 cm^{-1} .

3. THEORY AND DISCUSSION OF RESULTS

To describe the observed anomalies in the high-frequency magnetic properties of hematite we make the usual assumptions^{1,8–10} and use the Landau-Lifshitz equations with a thermodynamic potential in the form

$$\Phi(\mathbf{m}, \mathbf{l}) = \frac{1}{2} A m^2 + \frac{1}{2} K_1 l_z^2 + \frac{1}{4} K_2 l_z^4 - d(m_x l_y - m_y l_x) - M_0 \mathbf{m} \mathbf{H}, \quad (2)$$

where $\mathbf{m} = (\mathbf{M}_1 + \mathbf{M}_2)/2M_0$, $\mathbf{l} = (\mathbf{M}_1 - \mathbf{M}_2)/2M_0$, and \mathbf{M}_1 and \mathbf{M}_2 are the sublattice magnetizations ($M_1^2 = M_2^2 = M_0^2$).

A spontaneous first-order phase transition between the weakly ferromagnetic state ($l_z = 0$) and the antiferromagnetic state ($l_z = \pm 1$) occurs at $K_1^{\text{eff}}(T) + (1/2)K_2 = 0$, where $K_1^{\text{eff}}(T) = K_1(T) + d^2/A$, $K_2 < 0$.

Solving the linearized Landau-Lifshitz equations for small homogeneous oscillations about the equilibrium positions, we can find all the components of the high-frequency magnetic permeability tensors (see, e.g., Refs. 23 and 24). For $T > T_M$ the oscillations are excited in the quasiantiferromagnetic mode of interest to us by a high-frequency magnetic field $\mathbf{h} \parallel \mathbf{m}_0 \parallel \mathbf{H}_0$, where \mathbf{m}_0 is the spontaneous weak ferromagnetic moment whose direction is set in our experiments by the external magnetic field ($H_0 \sim 200\text{--}400$ Oe) in the basal plane. The oscillations $\Delta \mathbf{m}$ of the total magnetization are in this mode along \mathbf{m}_0 or \mathbf{h} : $\Delta \mathbf{m} = \chi(\omega) \mathbf{h}$. The corresponding component of the high-frequency magnetic-permeability tensor (neglecting H_0 is determined by expression (1) in which $\Delta \mu = 4\pi\chi(0)$, while

$$\chi(0) = \chi_{\perp} = M_0^2/A = M_0/2H_D$$

is the static susceptibility in the basal plane for the weakly ferromagnetic phase,

$$\nu_0 = (\gamma/2\pi) [2H_E H_{A1}^{\text{eff}}(T)]^{1/2}, \quad H_{A1}^{\text{eff}} = (K_1 + d^2/A)/M_0, \quad (3)$$

and γ is the gyromagnetic ratio. The damping in this mode is taken into account in (1) by the phenomenological coefficient Γ .

When we pass to the antiferromagnetic state ($T < T_M$) the frequency of the low-lying quasiferromagnetic mode increases discontinuously to the value of the frequency of the quasiantiferromagnetic mode, so that at H_0 the oscillations are degenerate, with a frequency

$$\nu_0 = (\gamma/2\pi) (2H_E |H_{A1}^{\text{eff}}(T) + H_{A2}|)^{1/2}, \quad (4)$$

where $H_{A2} = K_2/M_0$. The dynamic-magnetic-permeability components $\mu_{xx} = \mu_{yy}$ are likewise determined in this case by Eq. (1), in which ν_0 should be taken from (4), and the static susceptibility $\chi(0)$ is

$$\chi(0) = \chi_{\perp} + \chi_{\text{rot}}, \quad (5)$$

where the rotational susceptibility is

$$\chi_{\text{rot}} = \frac{m_0^2}{|K_1^{\text{eff}} + K_2|}, \quad m_0 = \gamma_{\perp} H_D, \quad H_D = d/M_0. \quad (6)$$

Equations (1) and (3)–(5) describe quantitatively the experimental results. We shall pay particular attention to the behavior of the dynamic susceptibility and, in particular, to the static contribution $\chi(0)$, which has a discontinuity at the transition point. The physical cause of this discontinuity is that the transition to the antiferromagnetic phase adds to the transverse susceptibility χ_{\perp} due to the shear of the magnetic moments also a rotational susceptibility χ_{rot} . The latter is due to the Dzyaloshinskii interaction in the crystal and tilts the antiferromagnetic moment relative to the threefold axis C_3 (the z axis) in a field $\mathbf{h} \parallel C_3$. Using the experimental values

$\Delta \mu = 0.00125$ at $T > T_M$, $\Delta \mu(T_M^-) = 0.002 - 0.0022$, and $\nu_0(T_M) = 2.8 \text{ cm}^{-1}$, we obtain with the aid of (3)–(5) for the transverse susceptibility and the Dzyaloshinskii field the estimates $\chi_{\perp} = 1.9 \cdot 10^{-5} \text{ cm}^3/\text{g}$ and $H_D = 22\text{--}25$ kOe. They agree well enough with the results static measurements, viz., $\chi_{\perp} = 1.8 \cdot 10^{-5} \text{ cm}^3/\text{g}$ (Refs. 1 and 7), $\chi_{\perp} = 1.95 \cdot 10^{-5} \text{ cm}^3/\text{g}$ (Refs. 5 and 9), and $H_D = 22.2\text{--}22.6$ kOe (Refs. 1, 5, 9).

The decrease of $\Delta \mu(T)$ when the temperature decreases below the discontinuity ($T < T_M$) (see Fig. 3b) is due to the decrease of χ_{rot} because of the increase of the anisotropy energy. It is interesting that the $\Delta \mu(T)$ temperature dependence is determined uniquely by the temperature dependence of the resonance frequency $\nu_0(T)$ and takes the form ($T < T_M$)

$$\Delta \mu(T) = 4\pi\chi_{\perp} [1 + \gamma^2 H_D^2 / (2\pi\nu_0(T))^2], \quad (7)$$

where χ_{\perp} and H_D are independent of T in this temperature interval.

Using the experimental $\nu_0(T)$ (Fig. 3a) and the values $H_D = 23$ kOe, $\chi_{\perp} = 1.90 \cdot 10^{-5} \text{ cm}^3/\text{g}$, and hematite density $\rho = 5.29 \text{ g/cm}^3$, we calculated $\Delta \mu(T)$ at $T < T_M$ shown by the solid line in Fig. 3b. It can be seen that on the whole it describes well the experimental $\Delta \mu(T)$, thus attesting to the self-consistency of the experimental results. Note also that the observed $\Delta \mu(T)$ agrees with the results of static measurements of the corresponding susceptibility of hematite for $T < T_M$ (Ref. 7). Good agreement between the experimental and calculated $\Delta \mu(T)$ obtains also for hematite doped with gallium at $\chi_{\perp} = 1.9 \cdot 10^{-5} \text{ cm}^3/\text{g}$, and $H_D = 20.5$ kOe, and for the experimental $\nu_0(T)$ (Fig. 3a).

We analyze now in greater detail the resulting temperature dependence of the AFMR frequency, $\nu_0(T)$, and ascertain in particular the extent to which current notions concerning the temperature dependence of the anisotropy constants in hematite describe $\nu_0(T)$. It is known^{25,2} that the $K_1(T)$ dependence in hematite is the result of two large but opposite contributions, viz., single-ion and dipole. Owing to their different temperature dependences, the quantity $K_1^{\text{eff}}(T) + (1/2)K_2(T)$ reverses sign, and it is this which leads to the Morin phase transition. Taking this into account, we represent the temperature dependence of the anisotropy constants, following the theory of Ref. 26, in the form

$$K_1^{\text{eff}}(T) = (K_1^{\text{dip}} + K_1^{\text{ex}}) l_0^2(T) + K_1^{\text{si}} \langle \bar{Y}_2^0 \rangle_T,$$

$$K_2(T) = K_2^{\text{si}} \langle \bar{Y}_4^0 \rangle_T, \quad (8)$$

where K_1^{dip} , K_1^{ex} , K_1^{si} are respectively the dipole, exchange ($K_1^{\text{ex}} = \chi_{\perp} H_D^2 \ll K_1^{\text{dip}}$) and single-ion contributions to the anisotropy constants at $T = 0$; $l_0(T)$ is the reduced magnetization of the sublattices;

$$\langle \bar{Y}_n^0 \rangle_T = \langle Y_n^0 \rangle_T / \langle Y_n^0 \rangle_0;$$

$\langle Y_n^0(S_z) \rangle_T$ are spherical harmonics averaged with a density matrix $\rho = \exp(S_z x)$;

$$x = (1/S) B_S^{-1}(l_0(T));$$

B_S^{-1} is the inverse of the Brillouin function, and $S = 5/2$ is the spin of the Fe^{3+} ion. We calculate $l_0(T)$ in the molecular-field approximation from the equation

$$l_0 = B_s(3ST_N l_0 / (S+1)T).$$

Using these values of χ_1 and H_D and the experimental AFMR frequencies at the Morin point and at $T=0$, we can determine from (3) and (4) the values $K_2(0) = K_2^{\text{si}} = -4.27 \cdot 10^4$ erg/g and $K_1^{\text{eff}}(0) = -6.2 \cdot 10^4$ erg/g. Note that they agree well with the corresponding hematite parameters obtained from static magnetic measurements.¹

Assuming now, according to the calculations of Ref. 15, that the dipole contribution is $K_1^{\text{dip}} = 3.512 \cdot 10^6$ erg/g ($H_{A1}^{\text{dip}} = K_1^{\text{dip}}/M_0 = 10.04$ kOe), we determine the dependence of ν_0 on temperature from Eqs. (3), (4), and (8). The corresponding $\nu_0(T)$ plot is shown in Fig. 3a (curve ●, $T_N = 900\text{K}$). Similar calculations for gallium-doped hematite at $K_2^{\text{si}} = -3.9 \cdot 10^4$ erg/g, $K_1^{\text{eff}}(0) = 1.13 \cdot 10^4$ erg/g, $K_1^{\text{dip}} = 3.336 \cdot 10^6$ erg/g, and $T_N = 900$ K yield the $\nu_0(T)$ dependence represented by the open circles in Fig. 3a. It can be seen from the figure that the agreement between the theoretical and experimental curves can on the whole be regarded as satisfactory especially at low temperatures. This attests to the validity of existing ideas concerning the mechanisms that determine the anisotropy energy and its temperature dependence in hematite. The more noticeable disparity of theory and experiment at high temperatures is apparently due to the larger error in the calculation of $l_0(T)$ in the molecular-field approximation.¹⁾ We note in this connection the high sensitivity of $\nu_0(T)$ and $K_1(T)$ to the values of $l_0(T)$, since $K_1(T)$ is determined by the competition between two contributions that are larger by approximately two orders than $K_1(T)$ and practically cancel each other.

We discuss now the nature of the anomalies observed in the temperature dependence of the transmission coefficient at $T = T_M$ (Fig. 2). Using our experimental values of ϵ and μ , we have calculated the temperature dependence of the transmissivity of our hematite sample of thickness $d = 1.232$ mm, using the known equations for the transmission of a plane-parallel layer. The experimental curves, which are the results of relative variation of the transmission coefficient with temperature, were tied to the corresponding theoretical curves at one point ($T = T_M$). As seen from Fig. 2, the experimental and calculated data are in good agreement. It is easy to verify that the magnitudes and signs of the discontinuities of the transmission-coefficient temperature dependence are due to a small relative frequency shift of the interference maxima in the transmission spectra above and below the transition, owing to the change produced in the refractive index $n = (\epsilon\mu)^{1/2}$ by the discontinuity $\Delta\mu$ at $T = T_M$.

Calculations of the $T(\nu)$ spectrum directly above and below the transition point show that the sign of the transmissivity discontinuity at the Morin point depends on the location of the measurement frequency relative to the interference maxima on the $T(\nu)$ plot.

We can thus conclude that the anomalies of the transmission-coefficient temperature dependence at $T = T_M$ are due to the discontinuity of the static magnetic permeability of the hematite at the Morin phase-transition point.

We conclude by discussing the behavior of the linewidth Γ . The hematite crystal we investigated had $\Gamma(300\text{K}) \sim 0.01\text{ cm}^{-1}$. Observation of AFMR in scanning

over the magnetic field (at constant ν) yields linewidths $\Delta H \sim 0.1\text{ kOe}$ for the low-frequency mode^{27,28} and $\Delta H \sim 1\text{ kOe}$ for the high-frequency one.²⁷ Using the known equations for the dependences of the AFMR frequencies on the magnetic field,^{23,24} it is easy to verify that these values of ΔH agree with the linewidth we observed. This indicates that the investigated crystals were all of like quality.

With respect to the temperature dependence of the linewidth $\Gamma(T)$ in $\alpha - \text{Fe}_2\text{O}_3$ and $\alpha - \text{Fe}_{2-x}\text{Ga}_x\text{O}_3$, we note that for $T < 400\text{K}$ it is apparently determined by extrinsic relaxation processes due to the presence of defects and impurities. At higher temperatures ($T > 400\text{K}$) the growth is due to the contribution of intrinsic (four-magnon, six-magnon, etc.) relaxation processes, a contribution that increases strongly with rise of temperature.²⁹

The largest maximum observed on the $\Gamma(T)$ plot in the region of the Morin temperature in both pure and Ga^{3+} -doped hematite, is apparently due to the some smearing ($\sim 1-2\text{K}$) of the transition by the inhomogeneities of the sample.

The increase of $\Gamma(T)$ in $\alpha - \text{Fe}_2\text{O}_3$ with decrease of temperature was unexpected. This behavior of $\Gamma(T)$ can be due to the presence of a small admixture of Fe^{2+} and Co^{2+} ions. Indeed, an investigation²⁸ of hematite with Co^{2+} impurity revealed an increase of the low-frequency mode linewidth at low temperatures. One cause of this increase, in the opinion of the authors of Ref. 28, is the freezing of the electronic $\text{Fe}^{2+} - \text{Fe}^{3+}$ and $\text{Co}^{2+} - \text{Fe}^{3+}$ transitions (see also Ref. 30). According to Clogston's theory³¹ (see also Ref. 32) this can lead at low temperatures to an increase of the linewidth and to the appearance of a maximum on the line.

4. CONCLUSION

We have thus obtained the temperature dependence of the parameters of the dynamic magnetic permeability of hematite and of hematite with Ga^{3+} impurity. Discontinuities were observed in $\Delta\mu(T)$ and $\Gamma(T)$ at the Morin point. Anomalous growth of $\Gamma(T)$ near liquid-helium temperatures was observed in hematite free of gallium.

The observed behavior of $\Delta\mu(T)$ was shown to be well described by a simple model that uses the Landau-Lifshitz equations with a thermodynamic potential that includes the first two anisotropy constants. Comparison of the behavior of the hematite and of hematite with Ga^{3+} impurity shows that introduction of the impurity is manifested mainly by a change of the $\nu_0(T)$ dependence, and hence also by the temperature dependence of the anisotropy energy; it furthermore changes insignificantly such quantities as $\Gamma(T)$ and the discontinuities of $\Delta\mu(T)$ at the Morin point. The comparison of the experimental and theoretical $\nu_0(T)$ dependences agrees on the whole with the contemporary notions concerning the mechanisms that determine the anisotropy energy, its temperature dependences in hematite, and the influence exerted on it by the Ga^{3+} impurities.

We have established that the anomalies in the temperature dependence of the transmission coefficient of hematite samples at the Morin point are caused by an abrupt change of the refractive index, due to a discontinuity of the magnetic permeability.

- ¹¹Our calculations have shown that the observed $\nu_0(T)$ dependence can be well described in the entire temperature range even in this approximation, but for larger (by 40–60%) contributions of K_1^{sl} and $K_1^{\text{dip}} + K_1^{\text{ex}}$. If K_1^{dip} remains unchanged this leads, however, to an unjustifiably strong increase of the exchange contribution K_1^{ex} compared with $\chi_1 H_D^2$.
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