ANTIFERROMAGNETIC RESONANCE IN HEMATITE IN THE WEAKLY FERROMAGNETIC STATE

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A detailed investigation is made of antiferromagnetic resonance in the weakly ferromagnetic phase of hematite (α-Fe_2O_3), corresponding to both the high- and the low-frequency branches of the spin-wave spectrum, over a wide range of frequencies (λ = 3.5 to 1.5 mm) and of temperature (T = 270 to 310°K). A strong temperature dependence of the high-frequency branch of the antiferromagnetic resonance is discovered and investigated. The temperature T_M of phase transition to a state without weak ferromagnetism is found; it agrees well with results obtained earlier. A large difference is found between the line widths of the high- and low-frequency resonances.

1. INTRODUCTION

As is well known, hematite (α-Fe_2O_3, space group D_{3d}^6) shows weak ferromagnetism at temperatures above $T_M \approx 250°K$[1-3]. The presence of weak ferromagnetism leads to the result that in one of the branches of the spin-wave spectrum, the energy gap in the absence of a magnetic field is quite small[4-6]. For this branch, the dependence of the antiferromagnetic resonance frequency on an external magnetic field applied in the basal plane, without allowance for anisotropy, has the following form[4-6]:

$$\omega_1 / \gamma = \sqrt{H(H + H_D) + H_{\Delta 1}^2}, \qquad (1)$$

where H_D = 22 kOe, $H_{\Delta_1}^2$ = 13.7 ± 0.4 kOe^2. The other branch of the spin waves, just as in the case of uniaxial antiferromagnets, has a quite large energy gap (the energy corresponds to a wavelength λ = 1.5 to 2 mm). Consequently, the antiferromagnetic resonance corresponding to this branch has hitherto not been studied in detail. There is a communication by Foner and Williamson[7] reporting that they observed the high-frequency branch in the neighborhood of the phase transition to the purely antiferromagnetic state ($T \gtrsim T_M \approx 250°K$) with the external magnetic field directed perpendicular to the basal plane; Elliston and Troup[8] also reported observation of antiferromagnetic resonance at a fixed frequency, when the external magnetic field was applied either in the basal plane or perpendicular to it. In these papers, however, there was no investigation of the dependence of the antiferromagnetic resonance frequency on the external magnetic field. It therefore seemed desirable to study the antiferromagnetic resonance that corresponds to the high-frequency spin-wave branch and that is described, in the case when the external magnetic field is applied in the basal plane[6,9], by the equation

$$\omega_2 / \gamma = \sqrt{H_c^2 + H_D(H + H_D)}, \qquad (2)$$

over a wide range of frequencies and temperatures. The size of H_c^2 is determined, as usual, by the effective exchange and anisotropy fields and also the magnetoelastic interaction. It also seemed useful to verify the validity of relation (1) in the submillimeter range of radio waves, in view of peculiarities recently discovered and not yet explained[10].

2. SPECIMENS AND EXPERIMENT

In our research we used synthetic monocrystals of hematite, grown in the Institute of Solid State Physics of the Czechoslovak Academy of Sciences[1)]. The specimens were in the form of thin plates of dimensions 1.5 × 0.5 × 0.2 mm. The plane of the plates coincided with the basal plane of the crystal. The measurements were made on a direct-amplification spectrometer that permitted operation over the range 1.4 to 3.5 mm. We used a transmission scheme with the specimen placed in a waveguide. As sources of electromagnetic radiation we used backward-wave tubes[11] [2)]. The wavelength of the radiation was measured with a Fabry-Perot interferometer with metal gratings[12]. The absorption signal was registered on a two-coordinate recorder. A voltage proportional to the size of the magnetic field was obtained with a film Hall-emf detector of indium arsenide; it was linear with the necessary accuracy over a wide range of magnetic fields[13] [3)]. The linearity of all the detectors used was specially verified by EPR in diphenylpicrylhydrazyl (DPPH) over the whole operating range of magnetic fields. As source of a stabilized current for the Hall detector, the apparatus described earlier[14] was used. The temperature of the specimen was measured with a thermocouple and was stabilized with an accuracy of ±0.1° over the interval 270 to 310°K by means of the system described in[15].

[1)]The authors are grateful to M. Vihr, of the staff of the Institute of Solid State Physics of the Czechoslovak Academy of Sciences for providing the monocrystals.

[2)]The authors are grateful to Professor M. B. Golant, who developed the tubes used in this work.

[3)]The indium arsenide Hall-emf detectors were kindly supplied by S. G. Shul'man of the staff of the Semicond. Inst. of the USSR Academy of Sciences.

3. RESULTS OF THE EXPERIMENT

Measurements were made both at a fixed temperature with varying frequencies, and at varying temperatures with a fixed frequency. In all experiments, the external magnetic field was directed parallel to the basal plane of the crystal. Absorption lines were observed that correspond both to the low-frequency (Fig. 1,a) and to the high-frequency (Fig. 1,b) branch. The hypothesis was confirmed that the position of the antiferromagnetic resonance line corresponding to the lower spin-wave branch does not depend on temperature within the temperature interval investigated. A strong temperature dependence was found for the position of the antiferromagnetic resonance line corresponding to the high-frequency spin-wave branch (Fig. 2). The temperature dependences of the positions of the antiferromagnetic resonance lines for various wavelengths are shown in Fig. 3.

It should be mentioned that both the shapes and the widths of the absorption lines of the high-frequency branch differed somewhat at different frequencies, whereas the absorption line of the low-frequency branch experienced practically no change either of its shape or of its width. But in view of the fact that the shapes and widths of the lines did not change very much for different temperatures, it seemed advisable to investigate the antiferromagnetic resonance for fixed frequencies and varying temperature.

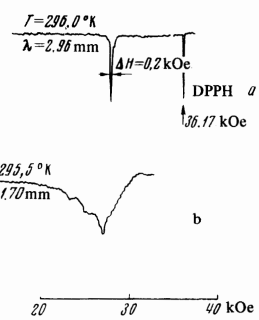

FIG. 1. Absorption line in antiferromagnetic resonance in hematite: a—resonance corresponding to the low-frequency branch of the spin-wave spectrum, and the field mark according to diphenylpicrylhydrazyl (DPPH); b—resonance corresponding to the high-frequency branch of the spin-wave spectrum.

4. DISCUSSION OF RESULTS

Hematite is among the most widely investigated antiferromagnets. Nevertheless, the experimental results obtained by different authors usually differ from each other; this is apparently a consequence of the different origins of the specimens. It should be mentioned that results obtained on natural and on synthetic monocrystals differ very greatly in high-frequency measurements[3], while in static measurements the difference is significantly smaller. The present investigation was made on the same hematite monocrystals, with a quite narrow absorption line in the low-frequency branch of the spectrum, as in preceding work[3,6]; this allows a valid comparison of the results obtained earlier with the new results.

As is known, the Hamiltonian for an antiferromagnet of α-Fe_2O_3 type, without allowance for anisotropy in the basal plane or for magnetoelastic and elastic terms, can be written in the following form:

$$\mathcal{H} = {}^1/_2 B\mathbf{m}^2 + {}^1/_2 \tilde{a} l_z^2 + q(l_x m_y - l_y m_x) - \mathbf{mh}.$$

The z axis is directed along the ternary axis of the crystal, perpendicular to the basal plane;

$$\mathbf{m} = (\mathbf{M}_1 + \mathbf{M}_2)\,/\,2M_0, \quad \mathbf{l} = (\mathbf{M}_1 - \mathbf{M}_2)\,/\,2M_0,$$

where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations ($M_1^2 = M_2^2 = M_0^2$), and $\mathbf{m}\cdot\mathbf{h} = 2M_0\mathbf{m}\cdot\mathbf{H}$ is the energy of the antiferromagnet in the external magnetic field (hereafter we shall always consider the external magnetic field to be applied in the basal plane). The sign of the anisotropy constant $\tilde{a}$ determines the magnetic state of the hematite; vanishing of $\tilde{a}$ with change of temperature determines the location of T_M in the absence of an external magnetic field (it should be mentioned that this is valid only without allowance for anisotropy constants of higher order, which are small)[2]. In the weakly ferromagnetic state, $\tilde{a} > 0$.

By considering small oscillations of $\mathbf{m}$ and $\mathbf{l}$ in the vicinity of the equilibrium values $\mathbf{m}_0$ and $\mathbf{l}_0$, one can obtain the characteristic frequencies of antiferromagnetic resonance

$$\omega_1 / \gamma = \sqrt{H(H + H_D)}, \tag{3}$$

$$\omega_2 / \gamma = \sqrt{\tilde{H}_c^2 + H_D(H + H_D)}, \tag{4}$$

where

$$\tilde{H}_c^2 = \tilde{a}B / 4M_0^2, \quad H_D = q / 2M_0.$$

Allowance for magnetoelastic interaction leads to relations (1) and (2). An experimental test of (1), by treatment of the results of the present measurements, together with data obtained earlier[3], by the method of

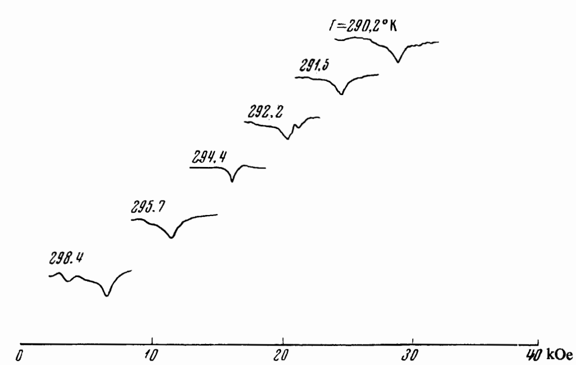

FIG. 2. Resonance absorption line in the high-frequency branch of spin waves for different temperatures; wavelength of the electromagnetic radiation, λ = 1.79 mm.

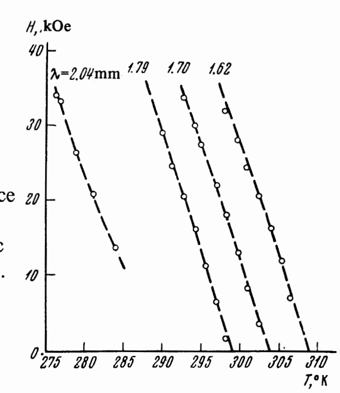

FIG. 3. Temperature dependence of the position of the line of the upper branch of antiferromagnetic resonance for various wavelengths.

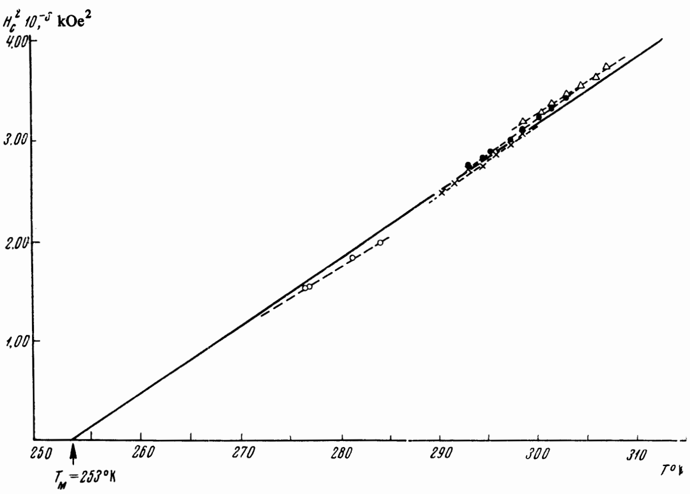

FIG. 4. Temperature dependence of H_c^2 for various wavelengths. Shown dashed are the dependences obtained at wavelengths: Δ, λ = 1.62 mm; ●, λ = 1.70 mm; X, λ = 1.79 mm; O, λ = 2.04 mm. The continuous curve shows the relation $H_c^2 = aT + b$, where $a = (0.067 \pm 0.005) \cdot 10^3$ kOe²/deg, $b = (-16.9 \pm 0.8) \cdot 10^3$ kOe².

least squares, leads to the values $H_D = 22$ kOe and $H_{\Delta 1}^2 = 13.7$ kOe² obtained in the cited paper, with errors not exceeding the limits indicated there. Figure 5, below, gives experimental results on antiferromagnetic resonance in the low-frequency branch (lower curve); the continuous line is described by the equation obtained by the method of least squares and given above.

In view of the quite good agreement of theory and experiment for the low-frequency branch of antiferromagnetic resonance, it was desirable to do an analysis of the experimental data on the high-frequency branch by use of relations (2) and (4). It should be mentioned that in relation (2), the magnetoelastic interaction enters implicitly in the value of H_c^2[6],

$$H_c^2(T) = \tilde{H}_c^2(T) + H_{\Delta 2}^2 = \frac{B}{4M_0^2}[\tilde{a}(T) + a_{\text{m.s.}}],$$

where $H_{\Delta 2}^2$ and $a_{\text{m.s}}$ are determined by the spontaneous striction and the mechanical stresses and therefore are independent of temperature. The values of H_c^2 obtained by means of relation (2) are shown in Fig. 4.

As is easily seen, values of H_c^2 can be obtained from the experimental data only by use of a quite accurately known value of H_D. Furthermore, the temperature dependence of $H_c^2(T)$ reflects, except for constant multipliers, the temperature dependence of the anisotropy constant $\tilde{a}(T) + a_{\text{m.s}}$ and should therefore be the same for measurements at all wavelengths. It is clear from Fig. 4 that there are four series of points, corresponding to the four wavelengths; and the points belonging to each series lie, with a good degree of accuracy, on straight lines of the form $H_c^2 = aT + b$. The equations of the straight lines corresponding to each series were obtained by the method of least squares. The graphs of these straight lines are shown dashed in Fig. 4. The values of a and b were calculated for each series. The scatter in the values of a and b (for a it amounted to ±7%, for b it was ±5%) can be explained if we suppose that a resonance absorption line consists of an unresolved group of lines, each of which is determined by resonance in different blocks of the monocrystal with different magnitudes and directions of the internal stresses, and that at different frequencies the chief contribution to the absorption line may come from different blocks[6,16]. The small changes of temperature that occurred in our measurements at fixed frequency did not significantly change the internal stresses in the specimen; there was also no change in the conditions of excitation either of the specimen as a whole or of its separate blocks. On change of temperature, therefore, the shape of the line did not change very much (Fig. 2); all the observed changes were presumably determined by the relation between $\tilde{a}(T)$ and $a_{\text{m.s}}$ for each block individually and for the specimen as a whole. It seemed desirable to obtain a mean value of a, determining the temperature dependence of the anisotropy in the neighborhood of the transition point investigated, and a mean value of b, the deviations from which, for each series of measurements, describe in order of magnitude the mean value of the mechanical stresses in the block. An averaging carried out thus leads to the following relation:

$$H_c^2(T) = aT + b,$$

$$a = (0.067 \pm 0.005) \cdot 10^3 \text{ kOe}^2/\text{deg} \quad b = (-16.9 \pm 0.8) \cdot 10^3 \text{ kOe}^2$$

Extrapolation of this relation to $H_c^2 = 0$ leads to the value $T_M = 253$°K, which differs by 3 to 4° from the value of T_M obtained earlier[3] from the vanishing of antiferromagnetic resonance in the low-frequency branch (if we take as the transition temperature T_M that temperature at which the intensity of the antiferromagnetic resonance decreases by one half; the whole transition range, for the crystals investigated, is according to the data of[3] ± 4°). Extrapolation to zero of

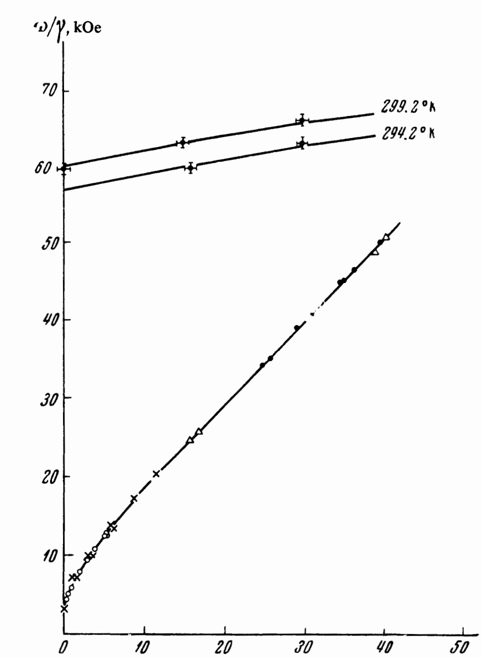

FIG. 5. Dependence of resonance frequencies, for the upper and lower branches of the spin-wave spectrum, on an external magnetic field applied in the basal plane: ●, data of the present research; O, data of reference [3]; Δ, [8]; X, [18].

each series of points also leads to a quite narrow transition range: ±8° from some mean value of T_M, differing little from that mentioned above.

We also remark that the antiferromagnetic-resonance line widths of the high-frequency and the low-frequency branches differ quite strongly (see, for example, Fig. 1) for all the frequencies and temperatures investigated (on a frequency scale, the ratio of line widths for the frequency range investigated amounts to $\Delta\omega_2/\Delta\omega_1 \sim \frac{1}{5}\Delta H_2/\Delta H_1$, where ΔH_2 and ΔH_1 are the absorption line widths for the high-frequency and the low-frequency branches, respectively). If we suppose that the block hypothesis mentioned above is right, we must recognize that the magnetoelastic constants entering only in the high-frequency branch (and which determined $H^2_{\Delta 2}$ and $H''_A(p)$ in the notation of[6]) are appreciably larger than the magnetoelastic interaction constants that enter in the value of the characteristic frequencies for the low-frequency branch of antiferromagnetic resonance. There is also another possibility: in the deformed blocks, stresses dominate which satisfy the condition $u_{xx} + u_{yy} = \Delta V/V - u_{zz} \neq 0$, and these have the greatest influence on the resonance in the high-frequency branch (deformations of the form $u_{xx} + u_{yy} \approx 0$, $u_{zz} \approx 0$ have a greater influence on resonance in the low-frequency branch; see[6,16]).

We note incidentally that the strong tendency—predicted in reference[17]—toward a change of antiferromagnetic-resonance line width in the low-frequency branch with change of frequency was not detected (the line width at wavelength 8 mm was $\Delta H \approx 200$ Oe[3], the line width at wavelength 3 mm was $\Delta H \approx 200$ Oe); this is presumably due to the fact that the observed line width is not caused by the mechanism discussed in the cited paper.

The averaged value of $H^2_c(T)$ obtained can be inserted in equation (2). The resulting relations for the two temperatures 299.2 and 294.2°K, and the experimental points corresponding to them, are shown in Fig. 5. As can be seen from this figure, the agreement between the theoretically calculated curves and the experimental points is satisfactory. We remark also that, apparently, the chief reason for the fact that the antiferromagnetic resonance corresponding to the high-frequency branch of the spin waves has not hitherto been investigated in detail is its strong temperature dependence, in consequence of which its investigation required good stabilization of the temperature over a wide range.

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