Mössbauer study of the critical behavior of hematite

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The Mössbauer spectra of a hematite (α -Fe₂O₃) single crystal are investigated near the Neel temperature in external magnetic fields from 0 to 23 kOe. The field-induced antiferromagnetic ordering predicted by Borovik-Romanov and Ozhogin is observed. The critical exponents and the amplitudes are determined in the temperature range $2 \times 10^{-4} \le |T - T_N|/T_N \le 2 \times 10^{-2}$ are $\beta = 0.365 \pm 0.011$, $B = 1.24 \pm 0.05$, $\delta = 4.4 \pm 0.1$, $D = 0.232 \pm 0.012$, $\gamma = \gamma' = 1.24 \pm 0.05$, $\Gamma' = (3.2 \pm 0.1) \times 10^{-4}$, and $\Gamma = (1.10 \pm 0.05) \times 10^{-3}$.

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INTRODUCTION

The study, by the method of Mössbauer spectroscopy, of the critical behavior of antiferromagnetic crystals with Dzyaloshinskii interaction makes it possible to obtain relatively simple information on the dependence of the order parameter on the temperature and the magnetic field.¹ This information is frequently difficult to obtain by other methods. One of the best known antiferromagnets of this type is hematite (α -Fe₂O₃), the magnetic properties of which have been investigated in great detail at low temperatures.² The magnetic behavior in the vicinity of the Néel point $T_N \approx 950$ K has not yet been sufficiently well studied. So far, only the critical exponents α and α' for the heat capacity³ and the exponent β for the magnetization⁴ have been obtained, the latter being determined at temperatures quite far from the critical point.

We deemed it therefore necessary to study in greater detail the magnetization of the hematite sublattices in the critical temperature region, with an aim not only at obtaining a more accurate value of β , but also of measuring the exponents δ for the magnetization and γ and γ' for the susceptibility. In addition, it is of interest to compare the results with the analogous data for iron borate FeBO₃, which is isomorphous to hematite, as well as with the results of theoretical calculations.

EXPERIMENT

The sample for the Mössbauer measurements was a single-crystal hematite plate measuring $10 \times 10 \times 0.08$ mm, grown at the Physics Institute of the Czechoslovak Academy of Sciences by E. Novak and P. Benes. The plane of the plate coincided with the basal plane (111).

The samples were placed in a high-temperature lowgradient furnace placed, for the measurements in an external field, between the poles of the electromagnet in such a way that the direction of the field H coincided with the (111) plane, and the direction $\mathbf{k}_{,}$ of the gamma quanta coincided with the C_3 axis. The Mössbauer measurements were made with an NOKIA LP-4840 multichannel analyzer, as well as with a single-channel spectrometer at constant velocities of the source (Co⁵⁷ in an Rh matrix). The instrumental width of the spectral lines was 0.3-0.5 mm/sec.

The sample temperature was measured with a Pt-Pt

(10% Rh) thermocouple and maintained constant within 0.1 K for 8–10 hours. The relative temperature drop along the sample did not exceed 3×10^{-4} at $T \approx 950$ K. The Néel point was determined by Mössbauer-spectroscopy temperature-scanning method and turned out to be $T_N = 951.2 \pm 0.1$ K.

RESULTS AND DISCUSSION

Hematite is a rhombohedral (space group D_{3d}^6) antiferromagnet with a weak ferromagnetic moment lying in the easy plane (111) at $T > T_M \approx 260$ K. According to Ref. 5, the effective uniaxial anisotropy field at room temperature is $H_A^{"} \approx 0.3$ kOe, whereas in the plane itself the anisotropy is low⁶ ($H_A^{"} \approx 1$ Oe).

Figure 1 shows the Mössbauer spectra of hematite in the vicinity of T_N . In the paramagnetic temperature region there is observed an incompletely resolved quadrupole doublet (spectrum 2) with a quadrupole splitting $\Delta E_Q = 0.43 \pm 0.01$ mm/sec. For most measured spectra this quantity was of the order of the Zeeman splitting ΔE_H of the levels of the nucleus. Therefore the effective fields H_{eff} at the nuclei were calculated with corrections for the additional shift of the sublevels of the excited state of the nucleus (I=3/2).⁷

Application of a field H = 23 kOe at $T = T_N$ induces at



FIG. 1. Mössbauer spectra of α -Fe₂O₃ near T_N (the C₃ axis coincides with the direction of k_γ , $H \parallel (111)$): curve 1—for H = 0, $T - T_N = 1.3$ K; 2—for H = 0, $T = T_N$; 3—for H = 22.9 kOe, $T = T_N$.

the nuclei an effective field $H_{eff} = 50 \pm 2$ kOe, thus attesting to a restoration of the antiferromagnetic order in the sample. The value of the hyperfine field $H_{hf}(T_N, H) = (H_{eff}^2 - H^2)^{1/2} = 44 \pm 2$ kOe allows us to calculate, within the framework of the phenomenological theory,^{8,9} the constant E_2 corresponding to the biquadratic exchange: $E_2(\alpha - Fe_2O_3) = 110 \pm 10$ kOe.

Figure 2 shows the isotherms of the magnetization M of the hematite sublattices. Here $\tilde{H}_{hf}(T,H) = H_{hf}(T,H)/H_{hf}(0,0) = \tilde{M}(T,H)$.

The critical parameters of α -Fe₂O₃ were determined from the $\tilde{H}_{\rm hf}(T,H)$ data using the formulas

$$\begin{split} \vec{M}(t,0) &= B(-t)^{s}, \ \vec{M} = M(t,H) / M(-1,0), \ \vec{M}(0,H) = D(\vec{H})^{1/\delta}, \\ \vec{H} &= q \mu_{B} S H / k T_{N}, \\ \chi(t,0) &= \frac{\partial \vec{M}}{\partial \vec{H}} \Big|_{T=0} = \begin{cases} \Gamma'(-t)^{-\gamma'}, & t < 0 \\ \Gamma(t)^{-\gamma'}, & t > 0 \end{cases}, \quad t = \frac{T - T_{N}}{T_{N}}, \end{split}$$
(1)

where β , δ , γ and γ' are the critical exponents; B, D, Γ , and Γ' are the critical amplitudes. The critical parameters were calculated by least squares. For the coexistence curve $\tilde{M}(t,0)$ we analyzed in addition the dependence of β on the upper limit t_m of the temperature interval in which the measurements were made. It was established that β stabilizes near a value $\beta = 0.365 \pm 0.011$ in the temperature interval $10^{-3} \leq |t| \leq 2 \cdot 10^{-2}$, in contrast to the value $\beta = 0.32$ in the interval $10^{-2} \leq |t| \leq 7 \cdot 10^{-1}$.⁴ Variation of T_N in the interval $t < t_m = 2 \cdot 10^{-2}$ yields a value $T_N = 951.25 \pm 0.16$ K, which agrees within the limits of errors with the value $T_N = 951.2 \pm 0.1$ K determined independently by the temperature scanning method.

The exponents $\delta = 4.4 \pm 0.3$, $\gamma' = 1.22 \pm 0.1$, $\gamma = 1.2 \pm 0.2$ determined from Eqs. (1) are subject to relatively large errors. To reduce the errors one can attempt to describe the entire family of isotherms with the aid of an equation of state that covers all the measured points in the (t, \tilde{H}) plane. According to similarity theory,¹⁰ the equations of state of a magnet in the critical region of temperatures and fields can be written in the form

$$h=f(m, t/|t|), h=\tilde{H}/|t|^{\beta\delta}, m=\tilde{M}/|t|^{\beta}.$$
(2)

The plot of the "scaling function" (Fig. 3), obtained from the experimental $\tilde{H}_{\rm hf}(T,\tilde{H})$ data and from the val-





FIG. 2. Magnetization isotherms of α -Fe₂O₃.

ues $\delta = 4.4$ and $\beta = 0.365$ shows that the experimental points lie on two branches that correspond to t > 0 and t < 0 and come together as $t \rightarrow 0$. According to Ref. 11 both branches can be approximated by the following power laws that satisfy the similarity-theory relations

$$h = b_0 m^{\delta} \pm b_1 m^{\delta - 1/\beta} + b_2 m^{\delta - 2/\beta} \pm \dots$$
(3)

near the critical isotherm $\tilde{M}(0,H)$ (the minus sign is used for t < 0) and

$$h = a_1 m + a_3 m^3 + a_3 m^5 + \dots$$
 (4)

near the curve $\tilde{\chi}(t,0), t>0$.

A least-squares reduction of the data has shown that within the limits of the experimental errors the first two terms of (3) describe well the branch *b* for all values of *m*, and the branch *a* for m > 0.9 at $b_0 = 617 \pm 18$, $b_1 = 1009 \pm 55$. The first three terms of (4) describe the branch *a* for $0 \le m \le 1.3$ at $a_1 = 909 \pm 27$, $a_3 = 96 \pm 3$, $a_5 = 580 \pm 60$.

The critical amplitudes obtained from the formulas $D = b_0^{-1/6}$, $\Gamma = a_1^{-1}$, $B = (b_1/b_0)^{\beta}$, $\Gamma' = \beta/b_0(b_1/b_0)^{-\gamma'}$, $\gamma' = \gamma = \beta$ ($\delta = 1$) have the following values: $D = 0.232 \pm 0.012$; $\Gamma = (1.10 \pm 0.07) \cdot 10^{-3}$, $\Gamma' = (3.2 \pm 0.1) \cdot 10^{-4}$; $B = 1.20 \pm 0.03$. The critical exponents δ , γ , and γ' are listed in the table.

According to the universality principle, the values of

FIG. 3. Plot of "scaling function" of hematite obtained from Mössbauer measurements: $a - at T > T_N$, $b - at T < T_N$. The solid lines correspond to Eq. (3) and the dashed line to Eq. (4).

TABLE I.

Substance	β	8	v'	v	Γ/Γ	$R_x = \Gamma D^{-\delta} B^{\delta-1}$	Refer- ences*
a-Fe ₂ O ₃ FeBO3	0.365±0.011 0.37±0.01	4.4±0.1 3.9±0.2	1.24±0.05 1.2±0.1	1.24 ± 0.05 1.2 ± 0.1	3.4±0.4	1.4±0.2 0.8±0.4	[PW] [¹]
Similarity theory, & expansion							
n=1 $n=2$ $n=3$	0.34 0.36 0.38	4.462 4.460 4.458		1.24 1.30 1.34	4.8 - -	$\left \begin{array}{c} 1.6\\ -\\ 1.33\end{array}\right\}$	[1 ²], [1 ³]

*[PW]-present work.

 $\beta, \delta, \gamma, \gamma', R_x = \Gamma D^{-6}B^{5-1}$ and Γ/Γ' depend mainly on the dimensionality *n* of the order parameter and on the dimensionality *d* of the lattice, should be the same for magnets belonging to one universality class (n, d).¹² Hematite and iron borate are easy-plane antiferromagnets, their critical behavior is therefore expected to be close to that of magnets of class (2, 3) (XY model).

It is seen from the table that the values of β , δ , and R_x for hematite are in good agreement with the theoretical values for n=2, although the values of γ for both substances are somewhat lower than the theoretical one. On the other hand, the exponents β , γ , and γ' for hematite agree within the limits of errors with the analogous exponents of iron borate. This also agrees with the assumptions that the two magnets belong to the same universality class.

The greatest difference between the critical parameters is observed for δ and R_x , which deviate for FeBO₃ towards the values predicted by the mean-field theory. This result can apparently be attributed to the fact that for hematite the interval of the reduced fields $2 \cdot 10^{-3} \leq \tilde{H} \leq 8 \cdot 10^{-3}$ in which the values of δ were determined lies somewhat closer to the critical point $(T_N, 0)$ than for iron borate, for which $10^{-2} \leq \tilde{H} \leq 3 \cdot 10^{-2}$.

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Thermal resistance of metal-insulator boundary and nonlinear electric resistivity of metal films at low temperatures

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The size effect in heat transfer and in the nonlinear electric resistivity of current-heated metallic plates (films) is discussed under conditions when the electronic contribution to the formation of the spectral distribution of the nonequilibrium ballistic phonons emitted by the metal plays the decisive role.

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1. INTRODUCTION

Experiments on the propagation of heat pulses in solids at high temperatures (see, e.g., Ref. 1) have increased recently the interest in the thermal resistance of a metal-insulator (M-I) interface. For an experimental study of this resistance (as well as to obtain the thermal pulses themselves), extensive use is made of electric current to heat thin metallic films deposited on bulky single-crystal insulator substrates. In these experiments one measures usually the magnitude and spectral intensity of the heat flux Q emitted by the film as a function of the electron temperature T_e and of the substrate temperature T (the latter can be regarded as equal to the temperature of the helium bath, inasmuch as the propagation of the phonons emitted by the film is ballistic). The electron temperature T_e is not measured directly but is calculated from the change of the film resistivity, using the known temperature dependence of the resistivity in the equilibrium case.²