Phase diagram of chromium in an external magnetic field

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The magnetic phase diagram of chromium is calculated in the magnetic field H-temperature T plane. It is shown that if the magnetic field deviates sufficiently from the direction of the wave vector of the spin-density wave, then the curve for the reorientational phase transition has an end point on the H–T plane. The coordinates of the critical point are calculated as functions of the angle of deviation of the magnetic field.

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1. A large number of papers[1,2] has been devoted to investigation of the magnetic properties of chromium. It is known[1] that after transition through the Neél temperature a transversely polarized, spin-density wave (SDW) corresponds to magnetic ordering in chromium. The **k** vector of the SDW is incommensurable with the lattice constant, but differs little from that value which corresponds to antiferromagnetic ordering. At the temperature T_{sf} there occurs a spin-reorientational phase transition, apparently of the first order, at which the transverse polarization of the SDW (relative to the **k** vector) changes to longitudinal polarization.[1] It is of interest to investigate the effect of the magnitude and direction of the external magnetic field on the spin-reorientational phase transition in chromium.

2. We shall construct the phase diagram on the basis of the following expression for the free energy

$$\Phi = B\mathrm{m}^2 - \mathrm{mH} + \frac{1}{2}\beta_1 |s_{\mathbf{k}}^z|^2 + \frac{1}{4}\beta^2 |s_{\mathbf{k}}^z|^4, \tag{1}$$

where m is the magnetization, s_k is the amplitude of the SDW, B is the exchange constant, and β_1 and β_2 are the first and second magnetic-anisotropy constants (the anisotropy z axis is parallel to the **k** vector). In writing expression (1) we took into account that as a result of transition to the magnetically ordered state the chromium lattice changes from a bcc lattice to a rhombic lattice which is very similar to a tetragonal lattice.[3] The magnetic anisotropy is produced due to the fact that the **k** vector eliminates the anisotropy axis. We ignore the change in direction of the **k** vector in the lattice because the reorientation of the **k** vector of the SDW is associated with a specific rearrangement of the Fermi surface, i.e., it requires an energy of the order of the electronic energies. The energy of the magnetic anisotropy, however, is much smaller than that of the electronic anisotropy. For simplicity, we ignore in the anisotropy energy the small terms associated with magnetization. Allowance for them produces small corrections for the results obtained, but greatly complicates the calculations. We also ignore the paraprocess, since the polarization amplitude of the SDW, as is well known,[4] is almost independent of temperature in the temperature range $T < T_{sf}$ ($T_{sf} = 123.5$ K, $T_N = 311$ K). Because of this, we ignore the contribution to

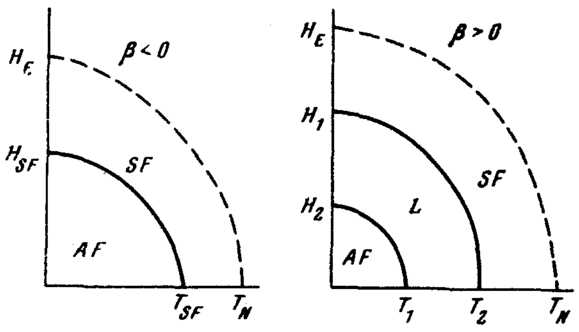

FIG. 1

the free energy from the entropy of the spin system and from terms such as $\mathbf{m}^2(\mathbf{s}_k\mathbf{s}_{-k})$ and $(\mathbf{m}\mathbf{s}_k)$ $(\mathbf{m}\mathbf{s}_{-k})$.

3. Minimizing Eq. (1) with allowance for the orthogonality condition on the $\mathbf{m}$ and $\mathbf{s}_k$ vectors, we can determine the main states of the system and the curves for the phase transitions. We present the results. If the magnetic field H is parallel to the z axis, then $\mathbf{m} = \chi_\perp H \sin\theta$ and the following states are stable:

$$\begin{aligned} &1.\ \theta_1 = 0 && \text{if } \beta_1 |s_k|^2 + \beta_2 |s_k|^4 + \chi_\perp H^2 \leq 0 \\ &2.\ \theta_2 = \pi/2 && \text{if } \beta_2 |s_k|^2 + \chi_\perp H^2 \geq 0 \\ &3.\ \cos^2\theta_3 = -(\beta_1 |s_k|^2 + \chi_\perp H^2)/\beta_2 |s_k|^4 && \text{if } \beta_2 > 0, \end{aligned} \tag{2}$$

where $\chi_\perp = 1/2\,B$ and θ is the angle between the antiferromagnetism vector $\mathbf{s}_k$ and the anisotropy axis. It can be seen from the given set of solutions that at $\beta_2 < 0$ two states are stable $\theta_1 = 0$ and $\theta_2 = \pi/2$. The curve for the phase transition can be determined from the condition $\Phi_1 = \Phi_2$, which has the form

$$\beta + \chi_\perp H^2 = 0\,; \quad \beta \equiv \beta_1 |s_k|^2 + \frac{1}{2}\beta_2 |s_k|^4 \tag{3}$$

or

$$H = H_{sf} = \sqrt{|\beta| / \chi_\perp}. \tag{3a}$$

In the absence of a magnetic field at $T = T_{sf}$ the H_{sf} field vanishes; hence $\beta = 0$ when $T = T_{sf}$. After expanding β in a series in powers of $T - T_{sf}$ in the neighborhood of T_{sf}, we rewrite Eq. (3) in the form.[1)]

$$T_{sf}(H) = T_{sf}(0)[1 - \chi_\perp H^2/\beta' T_{sf}(0)], \tag{4}$$

where

$$\beta' = \left(\frac{d\beta}{dT}\right)_{T = T_{sf}}; \qquad \beta(T_{sf}) = 0.$$

This result coincides with the field dependence of the temperature of the spin-

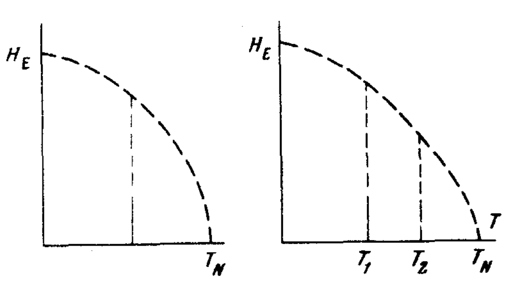

FIG. 2.

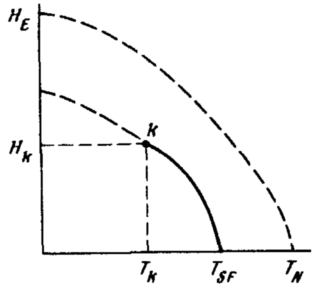

FIG. 3

reorientational transition obtained by Street *et al.*[1] using a different method. Equation (3a) determines the spin-flip fields at a specified temperature.

If the constant $\beta_2 > 0$, then the three phases are stable; the transition boundaries between them can be determined from the conditions

$$\beta_2 |s_k|^4 = -(\beta_1 |s_k|^2 + \chi_\perp H^2) \quad \text{for the transition} \quad \Phi_1 \rightleftarrows \Phi_3 \tag{5}$$

and

$$\beta_1 |s_k|^2 + \chi_\perp H^2 = 0 \quad \text{for the transition} \quad \Phi_2 \rightleftarrows \Phi_3 \,. \tag{6}$$

As can easily be seen, these two phase transitions are second-order. The order of the symmetry axis changes as a result of the $\Phi_1 \rightleftarrows \Phi_3$ transition and the symmetry plane vanishes as a result of the $\Phi_2 \rightarrow \Phi_3$ transition. The states of the system, however, vary continuously during these transitions. The transition interval is determined by β_2.

Expanding β_1 in a series with respect to the temperature, we represent Eqs. (5) and (6) in the form

$$T_{13} = T_0 - (\beta_2 |s_k|^4 + \chi_\perp H^2)/\beta_1' ; \qquad T_{23} = T_0 - \chi_\perp H^2/\beta_1' , \tag{7}$$

where

$$\beta_1' = |s_k|^2 \left(\frac{d\beta_1}{dT} \right)_{T = T_0} \quad \text{and} \quad \beta_1(T_0) = 0.$$

The magnetic field perpendicular to the anisotropy axis in a model with energy (1) does not affect the temperature of the spin-reorientational transition.

As mentioned above, in obtaining results (2)–(7), we ignored the paraprocess. Therefore, they are valid if $T < 1/2\ T_N$ and $H \ll H_E$ ($H_E = k_B T_N / \mu_0$, where k_B is the Boltzmann constant and μ_0 is the Bohr magneton). It is easy to see that these conditions are satisfied for the curves for the phase transitions.

4. The phase diagrams in the H–T plane are shown schematically in Figs. 1 and 2. As a result of changing the angle between the external magnetic field and the anisotropy axis, the phase diagrams in Figs. 1 are converted to the phase diagrams in Fig. 2.

The behavior of the phase point on the H axis in case of spin-flip of the first-order phase transition as a result of deviation of the magnetic field from the anisotropy axis was examined in Refs. 5 and 6. It was shown that there is a certain critical angle $\psi_{\rm crit}$ between the magnetic field and the anisotropy axis and at $\psi > \psi_{\rm crit}$ the phase transition is missing. These results give reason to assume that in our case the curves for the phase transition, beginning at certain angles $\psi_{\rm crit}$, have an end point (points), which is shown schematically in Fig. 3.

On the basis of expression (1) we can determine the "trajectory" of motion of the point "K" on the H–T plane by changing the angle ψ. If $\psi_{\rm crit}$ (0)$\leqslant\psi\leqslant\pi/8$, then the $H_k(\psi)$, $T_k(\psi)$ dependence is determined by the formulas:

$$\beta(T) = \frac{1}{2}\,\beta_2|s_k|^2\cot 2\psi\,; \qquad \chi_\perp H_k^2\sin 2\psi = \frac{\beta_2|s_k|^2}{2}\,; \quad \tan 2\psi_c\,(0) = \left.\frac{\beta_2|s_k|^2}{2\beta}\right|_{T=0} \tag{8}$$

To determine $T_k(\psi)$ we must know the $\beta(T)$ dependence. In the neighborhood of T_{sf} we have

$$T_k(\psi) = T_{sf} + \frac{\cot 2\psi}{2\beta'\beta_2|s_k|^2}\,; \qquad H_k^2(\psi) = \frac{\beta_2|s_k|^2}{2\chi_\perp\sin 2\psi}\,. \tag{8'}$$

This work was done during the author's stay in the Physics Department of the University of Toronto. Professor Fawcett has informed the author that the experiments on absorption of ultrasound in a chromium single crystal, which were performed by him and his co-workers, have confirmed the picture proposed in this paper that orientational phase transitions occur in chromium, depending on the magnitude and direction of the magnetic field. In particular, Fawcett and co-workers located the end point of the curve for the phase transition on the H–T plane.

The author thanks Professor Fawcett for many productive discussions and for showing the results of an experimental study of the phase diagram of chromium prior to its publication and Yablonski, Feder, and Lorentz for a discussion of the results.

[1)]In accordance with the usual representations, we assume that $\beta_2 = $ const and take into account the temperature dependence of only the β_1 constant.

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