

THE EFFECT OF UNIAXIAL COMPRESSION ON THE CRITICAL FIELD OF THE "SPIN FLIP" TRANSITION IN Cr₂O₃

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A sharp dependence of the anisotropy energy of antiferromagnetic Cr₂O₃ on pressure has been found. The effect cannot get a trivial explanation by features of magneto-dipole interaction. It may be connected with the properties of the spin-orbit interaction in the crystal.

Of transition metal ion sesquioxides isomorphous in crystal structure with corundum the uniaxial antiferromagnetic Cr₂O₃ is the subject of close attention caused by the specific properties of its magnetic symmetry which allows the existence of a magneto-electric effect. Its anisotropy energy *K* has been measured [1], but its nature is not understood yet finally. It has been found [2] that about half *K* arises from a magneto-dipole interaction. The present letter reports the anisotropy energy to be so sensitive to lattice geometry, that the crystal prostration along the rhombohedral axis C₃ with the external stress ~7kbar would produce a zero anisotropy energy and spontaneous "spin-over" of the spins into the basal plane. This result has been obtained while studying the effect of uniaxial compression along the C₃ axis on the critical field of "spin flip" transition in Cr₂O₃. The measurements have been carried out at 20.4°K in a pulsed magnetic field oriented along the C₃ axis to within 5' using a pair of balanced pickup coils [3]. The dependence of the critical field on the applied stress is shown in fig. 1. Within the experimental errors it may be described by a straight line whose slope gives

$$\frac{1}{H_c(0)} \frac{dH_c}{dp} = 7.5 \times 10^{-2} \text{ kbar}^{-1}$$

The value of critical field *H_c* and its dependence on the stress is given by *K* and the susceptibility difference $\Delta\chi = \chi_{\perp} - \chi_{\parallel} \approx \chi_{\perp}$:

$$H_c = (2K/\Delta\chi)^{\frac{1}{2}}, \quad \frac{1}{H_c} \frac{dH_c}{dp} = \frac{1}{2} \left(\frac{1}{K} \frac{dK}{dp} - \frac{1}{\Delta\chi} \frac{d(\Delta\chi)}{dp} \right)$$

The order of magnitude of the second summand may be got from the pressure dependence of the

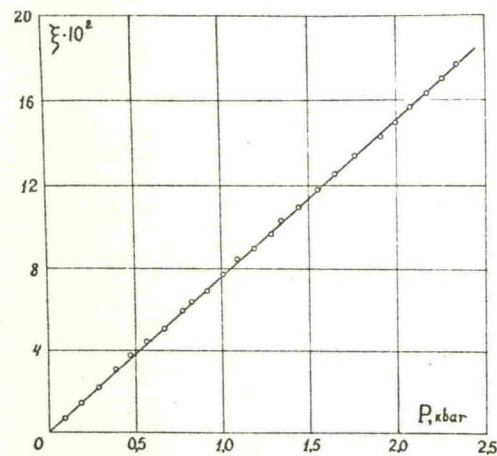


Fig. 1. Changes in the critical field of the "spin-flip" transition in Cr₂O₃ ($\xi = \{H_c(p) - H_c(0)\} / H_c(0)$) under uniaxial pressure along the rhombohedral axis. *T* = 20.4°K.

Néel temperature since both *T_N* and χ_{\perp} describe the superexchange interaction value in the crystal. However, the pressure effect on *T_N* [4] appears to be too small ($dT_N/T_N dp = -5 \times 10^{-3} \text{ kbar}^{-1}$) and consequently the value of the effect observed is accounted for by the anisotropy constant. Taking *K* = 2 × 10⁵ erg/cm³ [1] we find $dK/dp = 3.0 \times 10^{-5}$. It is interesting to note that the shift of *H_c* with pressure appeared to be by an order of magnitude larger than the corresponding one for Fe₂O₃, which was calculated from the magnetostriction jumps at the critical point [5]. Having the same crystalline structure and similar anisotropy energy and critical field values, Fe₂O₃ differs from Cr₂O₃ in the metal

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ion distribution over the magnetic sublattices. The difference in electronic shell structure of metal ions of these substances (ground states $6S_5$ and $4F_3$) seems to be important too. While the dependence of Fe_2O_3 may be accounted for by the magneto-dipole interaction [5], the corresponding calculations for Cr_2O_3 predict a value [2] which is by an order of magnitude less than the observed one.

It can be supposed that the effect observed may be accounted for by the anisotropic energy contribution connected with the spin-orbit interaction in the crystal. The present theory is unable to produce a quantitative estimate. However, one may expect that in a crystal with a large superexchange interaction ($T_N = 308^{\circ}K$) the properties of its anisotropic part [6] are significant in par-

ticular. They are noticeable against the small value of total anisotropy energy.

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EFFECT OF ELECTRON CORRELATIONS ON INTERACTION OF LOCALIZED MAGNETIC MOMENTS

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It is shown that the effect of electron correlations is to change the criterion for occurrence of localized magnetic states. The conditions of existence are different for parallel and antiparallel configurations of a pair of localized magnetic states. In general, the parallel configuration is favoured.

We consider the Anderson [1,2] model of two impurity transition metal atoms, each having a single, non-degenerate d-state, introduced into a matrix of a non-magnetic metal. The system of conduction and d-electrons is determined by the Hamiltonian (the notation of ref. 2 is used throughout).

$$\mathcal{H} = \sum_{k\sigma} \epsilon_k n_{k\sigma} + \sum_{i\sigma} E_0 n_{i\sigma} + \sum_{i\sigma} \frac{1}{2} U n_{i\sigma} n_{i,-\sigma} + \sum_{ik\sigma} V_{ik} C_{i\sigma}^+ C_{k\sigma} + \sum_{i\sigma} V_{12} C_{1\sigma}^+ C_{2\sigma} + \text{c.c.} \quad (1)$$

$C_{k\sigma}$, $C_{k\sigma}^+$ refer to the conduction electrons, $C_{i\sigma}$, $C_{i\sigma}^+$ correspond to the localized d-electrons and $n_{k\sigma} = C_{k\sigma}^+ C_{k\sigma}$, $n_{i\sigma} = C_{i\sigma}^+ C_{i\sigma}$. ϵ_k is the energy of conduction electrons, E_0 denotes the energy of the localized d-state, U is the Coulomb interaction energy; V_{ik} and V_{12} are the matrix elements for the mixing interaction of electrons. We assume $U \gg |V_{ik}|, |V_{12}|$.

The expectation values $\langle n_{i\sigma} \rangle$ are calculated by the Green function method [4,3]. The chain of the equations of motion for the Green functions $\langle\langle C_{i\sigma}; C_{i\sigma}^+ \rangle\rangle$ is reduced to a closed system by neglecting the Green functions $\langle\langle C_{k,-\sigma}^+ C_{i\sigma} C_{i,-\sigma} \rangle\rangle$, $\langle\langle C_{i\sigma}^+ \rangle\rangle$ and $\langle\langle C_{k,-\sigma}^+ C_{i\sigma} C_{i,-\sigma}^+ \rangle\rangle$ (cf. [3]), and by the following decouplings:

$$\langle\langle C_{k\sigma} n_{i,-\sigma}; C_{i\sigma}^+ \rangle\rangle = \langle n_{i,-\sigma} \rangle \langle\langle C_{k\sigma}; C_{i\sigma}^+ \rangle\rangle,$$

$$\langle\langle C_{j\sigma} n_{i,-\sigma}; C_{i\sigma}^+ \rangle\rangle = \langle n_{i,-\sigma} \rangle \langle\langle C_{j\sigma}; C_{i\sigma}^+ \rangle\rangle \text{ for } j \neq i.$$

For $U \gg |V_{ik}|, |V_{12}|$ we find finally

$$\langle n_{i\sigma} \rangle = \frac{1 - \langle n_{i,-\sigma} \rangle}{\pi} \times \left[\arctan \frac{E_F - E_0 - (1 - \langle n_{i,-\sigma} \rangle) \left(\lambda - \frac{|V_{12}|^2}{U \langle n_{j,-\sigma} \rangle} \right)}{\Delta (1 - \langle n_{i,-\sigma} \rangle)} + \frac{1}{2} \pi \right] \quad (2)$$

