Orthorhombic Anisotropy of Magnetic Susceptibility in Antiferromagnetic Chromium

M. O. Steinitz and E. Fawcett*
Department of Physics, University of Toronto, Toronto, Ontario, Canada

and

C. E. Burleson, J. A. Schaefer, L. O. Frishman, and J. A. Marcus†
Department of Physics, Northwestern University, Evanston, Illinois 60201
(Received 16 September 1971)

Measurements of the anisotropy of magnetic susceptibility in antiferromagnetic chromium show that the magnetic symmetry of a single-polarization domain in the transverse spin-density-wave phase is orthorhombic, indicating the existence of anisotropic terms in the susceptibility in addition to the spin anisotropy predicted by simple theoretical models.

We have measured the anisotropy of magnetic susceptibility in a single-$\vec{Q}$ crystal of chromium in both the transverse and longitudinal spin-density-wave (SDW) phases. In the transverse phase we apply a field perpendicular to $\vec{Q}$ large enough to produce an essentially single-polarization domain with its polarization direction $\vec{s}$ along the [100] axis perpendicular to both $\vec{Q}$ and $\vec{H}$. We then find the magnetic symmetry to be orthorhombic, with $\chi_Q - \chi_n$ and $\chi_n - \chi_s$, where $\vec{n} \perp \vec{Q} \perp \vec{s}$, the same order of magnitude. At lower fields, we observe a contribution to the anisotropy both from the growth of the single-polarization domain and from its intrinsic anisotropy. The data are interpreted using a thermal-activation model for polarization-domain growth. Comparison of the data with the Fedders-Martin[1] model shows the necessity for more detailed models for the antiferromagnetism of Cr, particularly the necessity for including anisotropy of effects such as the orbital paramagnetism and spin-orbit coupling.

Chromium has been shown to have a SDW antiferromagnetic structure[2] below $T_N = 312\,^\circ$K. The wave vector $\vec{Q}$ of the SDW lies along a [100] direction, and if a single crystal is cooled through T_N in a sufficiently large field $\vec{H}_c$ along a [100] direction, $\vec{Q}$ will lie parallel to $\vec{H}_c$ throughout the sample.[3] The single-$\vec{Q}$ state is preserved after $\vec{H}_c$ is removed, so long as the sample is not warmed through T_N. For $T_N > T > T_F = 122\,^\circ$K, the SDW is transversely polarized, while below T_F the SDW is longitudinally polarized.

In the transverse SDW phase, the ordered moments lie normal to $\vec{Q}$ and prefer to lie near [100] axes. The symmetry of a polarization domain is orthorhombic,[4] with the polarization direction $\vec{s}$ along a [100] direction perpendicular to $\vec{Q}$, the remaining [100] direction being denoted by $\vec{n}$. In general, one would expect an equal distribution of polarization domains between the two [100] axes normal to $\vec{Q}$ in the absence of an applied field. Since for any antiferromagnet the lowest-energy state is that with the ordered moments normal to an applied field (as $\chi_\perp > \chi_\parallel$), the application of a magnetic field $\vec{H}$ will tend to align the polarization with the [100] axis normal to $\vec{Q}$ along which the projection of $\vec{H}$ is smallest.[5,6] Below T_F, $\vec{Q}$ and $\vec{s}$ are parallel, giving tetragonal symmetry.

Rice[7] has shown that a Fedders-Martin–type model for chromium predicts that the spin susceptibility should behave like that of an ordinary antiferromagnet, i.e., for those electrons involved in the antiferromagnetism, the spin susceptibility approaches zero along the polarization direction $\vec{s}$ as the temperature approaches zero, but remains equal to the Pauli paramagnetic susceptibility in all directions normal to $\vec{s}$. Thus, the susceptibility of Cr along $\vec{Q}$ in the longitudinal phase (where $\vec{s}$ is parallel to $\vec{Q}$) should be reduced, giving an anisotropy in direct proportion to the decrease in the electronic specific heat between paramagnetic and antiferromagnetic Cr.[8,9] In the transverse phase, in an applied field large enough to align all the polarization domains into a single [100] direction $\vec{s}$, this model predicts that the susceptibility χ is isotropic in the plane normal to $\vec{s}$, that is, $\chi_Q = \chi_n$. The experimental results disagree with these predictions in both the transverse and longitudinal phases.

Our measurements were made on a single-$\vec{Q}$ crystal of mass 2.85 g, using a Condon torsion balance.[10] Similar measurements made by Montalvo[11,12] were inconclusive because of insufficient $\vec{Q}$ alignment in his crystals, since the highest cooling field available was 32 kOe. A previous magnetostriction experiment[4] on the same sample showed that the crystal used in these measurements, which we field cooled in $\vec{H}_c = 106$ kOe, would then be in a single-$\vec{Q}$ state and that at 127 °K a field of 22 kOe along one crystal axis perpendicular to $\vec{Q}$ aligns more than 90% of the domains along the third axis.[13] Curves of torque on the sample vs applied field $\vec{H}$ were taken with $\vec{Q}$ in the plane of $\vec{H}$ perpendicular to the suspension axis of the tor-

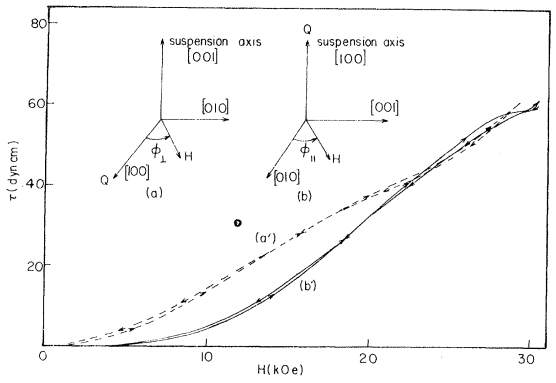

FIG. 1. Field dependence of magnetic torque in the transverse SDW phase of single-$\vec{Q}$ chromium at 129 °K (raw data). (a) Curve a': $\vec{Q} \perp$ suspension axis; $\phi_\perp = 45°$. (b) Curve b': $\vec{Q} \parallel$ suspension axis; $\phi_\parallel = 22\frac{1}{2}°$.

sion balance [Fig. 1(a)] and with $\vec{Q}$ parallel to the suspension axis [Fig. 1(b)]. The corresponding measured torques are shown in Fig. 1, curves a′ and b′, respectively. Since the polarization domains are more easily aligned at low temperatures, all field-dependence measurements in the transverse phase were made at $T = 129$ °K.

The curve of $\tau_\perp$ vs $\vec{H}$ (Fig. 1, curve a) is seen to have two distinct points of inflection. The first is due to the saturation of the number of polarization domains aligned parallel to [001]. The positive curvature of $\tau_\perp$ above the second point of inflection indicates that $\chi_Q - \chi_n$ is nonzero, since if χ were isotropic in the plane normal to $\vec{s}$ the torque curve would turn downward and approach zero as $\vec{H}$ is increased.

To interpret the torque quantitatively, we develop a model for polarization-domain orientation based on the thermal-activation model discussed by Werner, Arrott, and Kendrick.[14] In their model the number of domains having a given polarization is proportional to the Boltzmann factor $\exp[-E \times(\theta, \phi)\delta v/kT]$, where $E(\theta, \phi)$ is the sum of the crystal anisotropy and magnetic energy densities for a domain with polarization $\vec{s}(\theta, \phi)$, where ϕ specifies the field direction, θ specifies the polarization direction, and δv is the average volume of a domain. If one calculates the energy F of such a collection of domains, then $\tau = -\delta F/\delta\phi$ gives the torque on the sample. Our model[15] differs from that of Werner, Arrott, and Kendrick in two important respects: First, we assume that $\chi_Q - \chi_n$ is not identically zero, and second, we assume that the range of fields used in this experiment remains in the low-field region in which $\vec{s}$ always lies along a [100] direction.[16] Under this assumption, which is equivalent to the assumption that the magnetic energy density is much smaller than that of the crystal anisotropy, an exact solution for τ is possible for all orientations of $\vec{Q}$ with respect to $\vec{H}$. The question of whether $\vec{s}$ prefers to lie parallel to a [100] direction or perpendicular to $\vec{H}$ is irrelevant when $\vec{Q}$ lies in the plane of $\vec{H}$; since $\vec{s}$ must be perpendicular to $\vec{Q}$, the spins are driven along the third crystal axis, which is perpendicular to both $\vec{H}$ and $\vec{Q}$.

Using this model one obtains for the fraction of domains N having an energetically unfavorable polarization

$$N = \tfrac{1}{2}\{1 - \tanh[(\chi_n - \chi_s)H^2_{\text{eff}}\,\delta V/4kT]\}\,, \tag{1}$$

where H^2_{eff}, the square of the effective field felt by the spins, is given by

$$H^2_{\perp\text{eff}} = H^2 \sin^2\phi_\perp\,, \qquad H^2_{\parallel\text{eff}} = H^2 \cos 2\phi_\parallel\,. \tag{2}$$

The torques for the two orientations are then

$$\tau_\perp = -\tfrac{1}{2}mH^2 \sin 2\phi_\perp\{(\chi_Q - \chi_n) + \tfrac{1}{2}(\chi_n - \chi_s) \times[1 - f(\alpha H^2_{\perp\text{eff}})]\}\,, \tag{3}$$

$$\tau_\parallel = -\tfrac{1}{2}mH^2 \sin 2\phi_\parallel(\chi_n - \chi_s) f(\alpha H^2_{\parallel\text{eff}})\,,$$

where $f(\alpha H^2_{\text{eff}})$ is the distribution function for the polarization domains,

$$f(\alpha H^2_{\text{eff}}) = (\alpha H^2_{\text{eff}}) + \tanh(\alpha H^2_{\text{eff}}) - (\alpha H^2_{\text{eff}})\tanh^2(\alpha H^2_{\text{eff}}), \tag{4}$$

where $\alpha = (\chi_n - \chi_s)\delta v/4kT$ and m is the mass of the crystal.

In Fig. 2, we plot the data of Fig. 1 in terms of Eq. (3), showing the intercepts and asymptotes which we use to evaluate the susceptibility anisotropies $\chi_Q - \chi_n$ and $\chi_n - \chi_s$. The shapes of the experimental curves of Fig. 2 are in excellent agree-

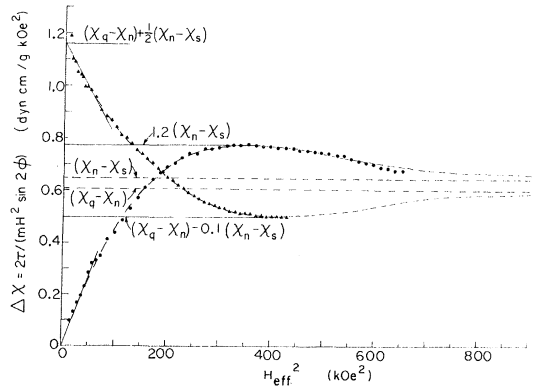

FIG. 2. Field dependence of anisotropy of susceptibility of Cr plotted in terms of the model discussed in the text. (a) Triangles: $\vec{Q} \perp$ suspension axis [see Fig. 1(a)]; (b) Circles: $\vec{Q} \parallel$ suspension axis [see Fig. 1(b)]. Dashed line, theoretical curve based on expressions (3) using measured values of $\chi_Q - \chi_n$, $\chi_n - \chi_s$, and α. H^2_{eff} defined in Eq. (2).

ment with that predicted by Eq. (4). Values of the sample-dependent parameter α, evaluated from the slopes, intercepts, extremals, and inflection points of Fig. 2 and Eq. (4), are consistent to within $\pm 10\%$ for both orientations, giving

$$\alpha = (0.31 \pm 0.03) \times 10^{-8} \text{ Oe}^2 .$$

The extremals of these plots should occur at the same value of H_{eff}^2; experimentally, they differ by about 15%. Three out of the four independent methods of determining α above are independent of the values of $\chi_n - \chi_s$ determined below.

Figure 2 shows clearly that $\chi_Q - \chi_n \neq 0$. From Fig. 2(a) we obtain the following values for the anisotropies of the susceptibility at $T = 129\,^\circ$K:

$$\chi_Q - \chi_n = 0.061 \times 10^{-6} \text{ emu/g} ,$$

$$\chi_n - \chi_s = 0.110 \times 10^{-6} \text{ emu/g} ,$$

and from Fig. 2(b) we obtain

$$\chi_n - \chi_s = 0.065 \times 10^{-6} \text{ emu/g} , \quad T = 129\,^\circ\text{K},$$

where emu/g = dyn cm/g Oe^2.

The discrepancy in the values of $\chi_n - \chi_s$ obtained from the two crystal orientations can be partially accounted for by spin pinning. Strains and imperfections in the crystal would be expected to pin polarization domains; such pinned domains would require a higher field to align. Before taking the data for $\tau_\perp$, the crystal was heated through T_F with $H = 29$ kOe along the crystal axis perpendicular to $\vec{Q}$, thus making it difficult for domains to become pinned in this direction. When the crystal was not so treated, the second inflection point was not clearly observed, demonstrating the existence of pinning effects. This treatment was not used in taking the data for $\tau_\parallel$. If 10% of the spin domains were pinned along each axis perpendicular to $\vec{Q}$, $\chi_n - \chi_s$ calculated from $\tau_\parallel$ would be 20% lower than the true value. In addition it should be noted that the value of $\chi_n - \chi_s$ obtained from Fig. 2(a) alone is a sensitive function of how one chooses the intercept.

From the values of α and $\chi_n - \chi_s$ given above, we calculate an average domain size

$$\delta v = 2.8 \times 10^{-16} \text{ cm}^3 \text{ to } 4.4 \times 10^{-16} \text{ cm}^3, \quad T = 129\,^\circ\text{K}.$$

This is in remarkably good agreement with an estimate by Munday and Street[16] of $\delta v = 1.4 \times 10^{-15}$ cm^3 using elasticity data and values of the anisotropy of strain between $\vec{s}$ and $\vec{n}$ from Ref. 10, as well as the estimate of the anisotropy energy of Werner, Arrott, and Atoji.[6] Moreover, two values of 4α reported by Werner, Arrott, and Atoji[6] at $T = 200$ and 295 °K are in good agreement with our value at 129 °K. A plot of $(\chi_n - \chi_s)\delta v H^2/k$ vs T for the three points suggests that the product $(\chi_n - \chi_s)\delta v$ is an increasing function of temperature.

The data seem to support the thermal-activation model discussed above rather than any simple form of the spin-pinning model such as that proposed by Bindloss[17] and applied to Cr by Werner, Arrott, and Atoji.[6] This model, depending on a harmonic restoring force on domain walls, predicts that the volume fraction of the crystal having spins along $\vec{H}$ should decrease as a linear function of H^2. τ/H^2 in Fig. 2(a) would then also decrease linearly with H^2. This is clearly not the case, as is shown in Fig. 2.

The temperature dependence of the anisotropy of susceptibility at 8 and 29 kOe is shown in Fig. 3. Below T_F the torque goes strictly as H^2. At 1.5 °K, the anisotropy of susceptibility is about 18×10^{-6} emu/mole. As the reduction in the density of states estimated from electronic specific-heat measurements of Heiniger[18] is about 1.5 mJ/mole °K^2, the anisotropy predicted from the assumption that χ_Q is reduced in direct proportion to this specific-heat reduction in the longitudinal phase, and that χ_n is unchanged from the Pauli paramagnetic susceptibility, is about 36×10^{-6} emu/mole. This difference is not explained by any of the other anisotropy components found here, as their magnitudes are all too small.

Two of us (E. F. and M. O. S.) are currently investigating the origins of the anisotropy by considering the contributions to the susceptibility from orbital paramagnetism and the effects of spin-orbit coupling on the spin paramagnetism. Shimizu[19] has discussed the anisotropy in the orbital paramagnetism. Rice[7] has pointed out the importance of including spin-orbit coupling effects in explaining the spin-flip transition. Such effects would also be expected to contribute to the anisotropy of susceptibility. Having modified the apparatus for large samples, two of us (C. E. B. and J. A. M.) are extending measurements of $\tau_\parallel$ and $\tau_\perp$ to three samples in fields up to 45 kOe.

We would like to acknowledge many helpful discussions with Professor Martin Zuckermann of

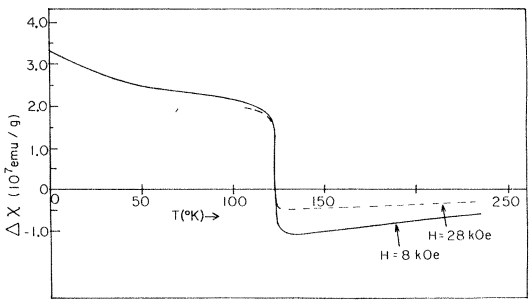

FIG. 3. Temperature dependence of the anisotropy of susceptibility of Cr for $\vec{Q}$ in the plane of $\vec{H}$ ($\vec{Q} \perp$ suspension axis).

McGill University. The authors at Toronto are grateful to the Materials Research Center at Northwestern University for the opportunity to perform these measurements.

*Work supported by the National Research Council of Canada.

†Work supported by the Advanced Research Projects Agency through the Northwestern University Materials Research Center and by the National Science Foundation.

[1]P. A. Fedders and P. C. Martin, Phys. Rev. 143, 245 (1965).

[2]A good review is provided in R. Street, B. C. Munday, B. Window, and I. R. Williams, J. Appl. Phys. 39, 1050 (1968).

[3]R. A. Montalvo and J. A. Marcus, Phys. Letters 8, 151 (1964).

[4]M. O. Steinitz, L. H. Schwartz, J. A. Marcus, E. Fawcett, and W. A. Reed, Phys. Rev. Letters 23, 979 (1969).

[5]S. A. Werner, A. Arrott, and M. Atoji, J. Appl. Phys. 39, 671 (1968).

[6]S. A. Werner, A. Arrott, and M. Atoji, J. Appl. Phys. 40, 1447 (1969).

[7]T. M. Rice, in Lectures at the Canadian Association of Physicists' Summer School, Banff, 1969 (unpublished).

[8]K. Andres, E. Bucher, J. P. Maita, and A. C. Sherwood, Phys. Rev. 178, 702 (1969).

[9]D. Bender and J. Müller, Physik Kondensierten Materie 10, 342 (1970).

[10]J. H. Condon and J. A. Marcus, Phys. Rev. 134, A446 (1964).

[11]R. A. Montalvo, Ph. D. thesis (Northwestern University, 1967) (unpublished).

[12]M. O. Steinitz and R. A. Montalvo, Bull. Am. Phys. Soc. 16, 381 (1971).

[13]In fact, it is more accurate to say that 90% of those domains which can be polarized in 100 kOe are polarized in 22 kOe. We cannot make any statement about regions of the crystal which cannot be polarized in 100 kOe and assume that these constitute a negligible fraction of the volume of the crystal, basing this assumption on neutron-diffraction experiments on other crystals (Ref. 5).

[14]S. A. Werner, A. Arrott, and H. Kendrick, Phys. Rev. 155, 528 (1967).

[15] C. E. Burleson, R. A. Montalvo, and J. A. Marcus, Bull. Am. Phys. Soc. 13, 58 (1968).

[16]B. C. Munday and R. Street, J. Phys. F 1, 498 (1971).

[17]W. Bindloss, J. Phys. Chem. Solids 29, 225 (1968).

[18]F. Heiniger, Physik Kondensierten Materie 5, 285 (1966).

[19]M. Shimizu, Progr. Theoret. Phys. (Kyoto) Suppl. 46, 310 (1970).

PHYSICAL REVIEW B VOLUME 5, NUMBER 9 1 MAY 1972

Deviations from Saturation and Phase Changes of the Magnetization in a Uniaxial Ferromagnet*

L. C. Bartel

Sandia Laboratories, Albuquerque, New Mexico 87115

(Received 22 September 1971)

A macroscopic Hamiltonian is used to describe and calculate the deviations from saturation and phase changes of the magnetization in a ferromagnet with a uniaxial magnetic anisotropy either parallel or perpendicular to an applied magnetic field. Using Green's-function techniques, we have solved for the component (time, spatially, and thermally averaged) of the magnetization in the field direction, and we have determined the static long-wavelength transverse and longitudinal susceptibilities (called the isolated susceptibilities). The stability limits, or critical fields, determined from the singularities in these susceptibilities occur under the conditions for which the elementary excitation energy vanishes. We found that the singularities in the transverse and longitudinal susceptibilities yielded the same stability limit. Under certain conditions (field applied along an easy axis of magnetization and for temperatures near the ordering temperature) the stability limit is determined by the singularity in the isothermal susceptibility. Results of the calculations for the magnetization as a function of applied field, at various temperatures and with various uniaxial anisotropies, are given.

I. INTRODUCTION

Spin-wave (SW) approaches have recently been used to investigate the deviation of the magnetization $\vec{M}$ from saturation (zero-point effects) for ferromagnets with a uniaxial anisotropy.[1,2] It was found that the net component of $\vec{M}$ along the applied-field direction became unstable when the long-wavelength SW energy vanished at a critical field. The micromagnetic approach has been used to determine the nucleation fields for various modes of deviation from saturation. In the usual forms of the micromagnetic approach,[3-5] it is assumed that M^2 is a constant. Recently, Minnaja[6] removed the re-