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Magnetic anisotropy due to spin-orbit and dipole-dipole interactions in spin-density-wave antiferromagnets

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The first effort to construct a theory of uniaxial magnetic anisotropy for an incommensurate spin-density-wave antiferromagnet like chromium is presented. The theory incorporates the spin-orbit and magnetic dipole-dipole interactions into a model for the magnetic ground state, using an idealized model band structure. The magnitude of the result is easily large enough to account for the uniaxial anisotropy of chromium. However, the temperature dependence of the anisotropy from both mechanisms is that of the antiferromagnetic gap squared. Therefore, a zero crossing of the anisotropy, as required to explain the spin-flip transition in chromium, does not occur. This indicates that for chromium the detailed band structure must be taken into account, or an additional mechanism is operative.

I. INTRODUCTION

Since the work of Brooks[1] in 1940 there has been a steady, if not large effort directed at obtaining a satisfactory theory of magnetic anisotropy for itinerant ferromagnets. Although this goal has not been entirely achieved, there has been demonstrable progress.[2] To date, however, there has been no effort to obtain a theory of magnetic anisotropy for itinerant antiferromagnets (AF). This is in spite of the fact that the spin-flip transition in chromium, elucidated experimentally nearly 15 years ago,[3] provides an intriguing challenge for such a theory.

Below the Néel temperature of 312 K, chromium displays antiferromagnetic order characterized by a linear spin-density wave (SDW) with an incommensurate $\vec{Q}$ vector.[4] As the temperature is lowered through the spin-flip temperature, $T_{sf} = 123.5$ K, the polarization of the SDW changes abruptly from transverse (AF1 phase) to longitudinal (AF2 phase). That fact that the magnetization, but not the $\vec{Q}$ vector, changes direction at T_{sf} suggests that in the single-$\vec{Q}$ state chromium possesses a uniaxial magnetic anisotropy with respect to the $\vec{Q}$ direction, which lies along one of the cubic axes of the paramagnetic state. This uniaxial anisotropy is in addition to a cubic anisotropy of the magnetization direction with respect to the crystalline axes, analogous to the cubic anisotropy found in ferromagnets having cubic symmetry in the paramagnetic state. The uniaxial anisotropy occurs because $\vec{Q}$ is incommensurate. If $\vec{Q}$ is commensurate, as in chromium alloyed with a small amount of manganese, a simply body-centered-cubic antiferromagnet results and this magnetic structure can be described equally well as a longitudinal SDW with $\vec{Q}$ along the direction of the magnetization or as one of two transverse SDW's with orthogonal $\vec{Q}$ vectors perpendicular to the magnetization. Thus the distinction between longitudinal and transverse cannot be made for the commensurate state and there is no spin-flip transition.

Many features of the spin-flip transition in chromium can be described[5] phenomenologically by assuming a uniaxial anisotropy term in the free energy

$$\Omega = K(T)(\hat{n} \cdot \hat{Q})^2, \tag{1}$$

where $\hat{n}$ and $\hat{Q}$ are unit vectors in the direction of the magnetization and $\vec{Q}$, respectively, and K is the temperature-dependent anisotropy constant. $K(T)$ must change from negative to positive as the temperature increases through T_{sf}. At $T=0$, K can be estimated from various experimental data to be about -2×10^4 erg/cm^3. Equation (1) could be augmented by the cubic anisotropy term, which has been experimentally demonstrated,[6] but it appears that the dominant anisotropy is the uniaxial one. Typically uniaxial or cubic anisotropy energies are the order of 10^5–10^6 erg/cm^3, so it can also be observed that even the dominant anisotropy of chromium is quite small. The phenomenological analysis also leads to the following expression for $K(T)$ in the temperature range 95 K$\leq T \leq T_{sf}$:

$$K(T) = -3.89 \times 10^4 (1 - T/T_{sf}) \text{ erg/cm}^3. \tag{2}$$

The theoretical challenge is thus to demonstrate a microscopic origin of the uniaxial anisotropy for an incommensurate SDW having first, the right magnitude and second, the proper temperature dependence. This paper describes the first attempt to accomplish these two goals. The procedure used is to incorporate the two common anisotropy mechanisms, the magnetic dipole-dipole interaction and spin-orbit coupling, into an idealized model of the magnetic ground state. The first goal of getting an adequate magnitude has been achieved, but the second goal of getting the right temperature dependence has not. The basic difficulty is that the temperature dependence of the anisotropy due to both mechanisms is the same, that of the antiferromagnetic gap squared, so that a zero crossing of the anisotropy coefficient does not occur. Theories of anisotropy for itinerant ferromagnets have also encountered difficulties in getting agreement with the experimental temperature dependence, and these difficulties are not yet completely resolved. The various suggestions for getting an improved temperature dependence in the ferromagnetic case do not appear to be directly applicable for chromium, as will be discussed later in the paper.

Before closing this introduction it is worth noting that the anisotropy energy in most magnetic materials is small compared to the energy of magnetic ordering. Therefore, the magnetic anisotropy is usually regarded as a perturbative feature of the magnetic ground state, and in that sense an anisotropy theory "comes after" a theory of the magnetic state. Nonetheless the magnetic anisotropy provides an important test of the model for the magnetic state in that the results of an anisotropy theory depend strongly on the basic model of the magnetic state. For example, models with localized spins lead generally to the rule that the temperature dependence of an anisotropy term of the lth order in the direction cosines of the magnetization is that of the $\frac{1}{2}l(l+1)$ power of the magnetization.[7] This rule is found to be well obeyed for most localized-spin magnetic insulators, but is not obeyed for the transition metals.

The remainder of this paper is organized as follows. Section II presents the calculation for the antiferromagnet and Sec. III discusses the results.

II. ANTIFERROMAGNET ANISOTROPY

The method of the anisotropy calculation, which is similar to that used for itinerant ferromagnets, is to model the magnetic state using an idealized band structure and then to determine the effect of the explicitly spin-dependent parts of the Hamiltonian, which are the one-electron spin-orbit term $\mathcal{H}_{so}$ and the two-electron magnetic dipole-dipole term $\mathcal{H}_{dd}$:

$$\mathcal{H}_s = \mathcal{H}_{so} + \mathcal{H}_{dd}, \tag{3}$$

where

$$\mathcal{H}_{so} = (1/2m^2c^2)\vec{s}_j \cdot [\vec{\nabla} \upsilon(\vec{r}_j) \times \vec{p}_j] \equiv \vec{\sigma}_j \cdot \vec{V} \tag{4}$$

and

$$\mathcal{H}_{dd} = \frac{e^2}{m^2c^2}\vec{s}_j \cdot \left(\frac{3(\vec{r}_j - \vec{r}_k)(\vec{r}_j - \vec{r}_k)}{r_{jk}^5} - \frac{1}{r_{jk}^3}\right) \cdot \vec{s}_k$$
$$\equiv \vec{\sigma}_j \cdot \overleftrightarrow{D} \cdot \vec{\sigma}_k, \tag{5}$$

with an obvious and standard notation. The idealized band structure adopted for the calculation is shown in Fig. 1. It is a nesting model as appears to be appropriate for chromium. The states near the Fermi level are orbital singlets, consistent with the findings of chromium band-structure calculations for the nesting states near E_F.[8] For $\vec{Q}$ incommensurate, states at $\vec{k}+\vec{Q}$ and $\vec{k}-\vec{Q}$ are inequivalent so three nesting bands crossing the Fermi level, $a\vec{k}$, $b+ = b\vec{k}+\vec{Q}$, and $b- = b\vec{k}-\vec{Q}$ must be included. These bands are augmented by excited states $c\vec{k}$, $d+ = d\vec{k}+\vec{Q}$, and $d- = d\vec{k}-\vec{Q}$ to treat $\mathcal{H}_{so}$, since $\mathcal{H}_{so}$ is diagonal in $\vec{k}$ and has vanishing expectation value for orbital singlets. Thus the six-band model has the minimum complexity for the situation at hand. The scheme of the calculation is to determine the six-band Green's function for the magnetic state, neglecting $\mathcal{H}_s$, in the Hartree-Fock (HF) approximation, and then to calculate corrections to the free energy due to $\mathcal{H}_{so}$ and $\mathcal{H}_{dd}$ with the six-band Green's function regarded as "unperturbed." Since the chromium anisotropy energy is small, a perturbative approach to treating $\mathcal{H}_s$ has been adopted.

Young and Sokoloff[9] (YS) have treated in the HF approximation the model with states a, $b+$, $b-$ in their discussion of the incommensurate ground state. In their calculation, YS define and include the Coulomb matrix elements of exchange (v) and direct (U) type, labeled by band indices as $v_{b-b-b+b+}$, v_{aab-b-}, v_{ab+b-c}, and $U_{b-b+b-b+}$. If Coulomb matrix elements involving the excited states are limited to $v_{d-b-d+b+}$, v_{cb+d-a}, v_{cad-b-}, and $U_{d-d+b-b+}$, extensive use can be made of the results of YS, because the HF equation of motion for the one-electron Green's function takes the block diagonal form (ω_ν is the frequency variable of a temperature-dependent Green's function)

$$\mathcal{G}^{-1}(\vec{k}, i\omega_\nu) = \begin{pmatrix} G_1^{-1} & 0 \\ 0 & G_2^{-1} \end{pmatrix}, \tag{6}$$

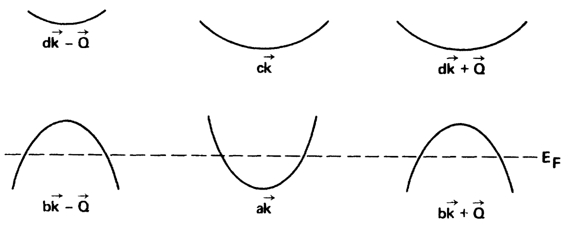

FIG. 1. Idealized band-structure model.

with G_1^{-1} being identical to the three-band $\mathcal{G}^{-1}$ of Eq. (10) in YS, and G_2^{-1} having the same form with the substitutions $a \to c$, $b- \to d-$, and $b+ \to d+$. The zeros of Eq. (6) neglect couplings only between states with dissimilar energies, but the important couplings of states of similar energy are included. There are six equations relating the nonzero off-diagonal matrix elements in Eq. (6) to matrix elements of $\mathcal{G}$, analogous to the three equations of Eq. (11) in YS.

Since $\mathcal{G}^{-1}$ is block diagonal, it can be inverted to find $\mathcal{G}$ exactly as done in YS, and the results inserted in the equations for the matrix elements to yield gap equations. YS found that for $\vec{Q}$ incommensurate, the ground state has simultaneously an SDW and CDW (charge-density wave) characterized by gaps denoted g and δ, respectively. In the present case there are also excited state SDW and CDW gaps, h and γ, and four coupled equations for g, δ, h, γ. If it is assumed that the gap magnitudes and kT are both much smaller than the average excited-state–nesting-state energy difference, to be denoted E_g, then these equations reduce to the two for g and δ given in Eq. (15) of YS and two which give h and γ in terms of g and δ:

$$h = (v_2/v_1)g, \quad \gamma = (v_2'/v_1')\delta, \tag{7}$$

with

$$\begin{aligned} v_1 &= v_{aab-b-} + v_{ab+b-a}, \\ v_1' &= v_{b-b-b+b+} - 2U_{b-b+b-b+}, \\ v_2 &= v_{cb+d-a} + v_{cad-b-}, \\ v_2' &= v_{d-b-d+b+} - 2U_{d-d+b-b+}. \end{aligned} \tag{8}$$

In this approximation the unoccupied excited states have little influence on the ground state, but are polarized by the ground state. In YS it is shown that the gap equations for g and δ can be solved for a specific model band structure. Therefore g, h, δ, γ can be regarded as known and so the (previously) inverted Eq. (6) gives the six-band Green's function. It is found that $\mathcal{G}^{ab+}$, $\mathcal{G}^{ab-}$, $\mathcal{G}^{cd+}$, and $\mathcal{G}^{cd-}$ are proportional to $\hat{n}\cdot\vec{\sigma}$ (SDW), while $\mathcal{G}^{b+b-}$ and $\mathcal{G}^{d+d-}$ are proportional to the spin identity (CDW).

The perturbative calculation to find the anisotropy energy due to $\mathcal{H}_{so}$ of Eq. (4) is straightforward. The matrix elements of the $\vec{V}$ of Eq. (4) are defined by

$$\begin{aligned} \langle a\vec{k}|\vec{V}|c\vec{k}\rangle &= \vec{V}^{ac}, \\ \langle b\vec{k}+\vec{Q}|\vec{V}|d\vec{k}+\vec{Q}\rangle &= \vec{V}^{bd+}, \\ \langle b\vec{k}-\vec{Q}|\vec{V}|d\vec{k}-\vec{Q}\rangle &= \vec{V}^{bd-}. \end{aligned} \tag{9}$$

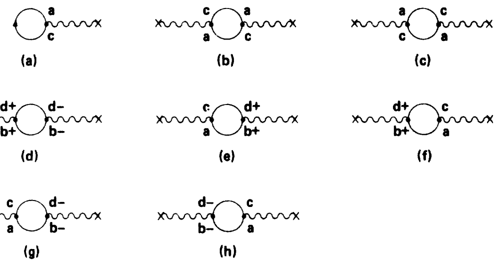

FIG. 2. Free-energy perturbation diagrams due to spin-orbit coupling.

Various first- and second-order diagrams that might contribute to the free energy are shown in Fig. 2. The perturbation $\vec{\sigma}\cdot\vec{V}$ is shown as an external field since it is a one-electron operator, and the trace of the spin operators around a fermion loop must be computed. The first-order diagram 2(a) is zero because $\mathcal{G}^{ac} = 0$ and because $\mathrm{Tr}(\overleftrightarrow{1}\sigma\cdot\vec{V}^{ac}) = 0$, and similarly for the second-order diagram 2(b). Diagram 2(c) is nonzero, but isotropic because $\mathrm{Tr}[(\vec{\sigma}\cdot\vec{V}^{ac})\overleftrightarrow{1}(\vec{\sigma}\cdot\vec{V}^{ac*})\overleftrightarrow{1}] \sim \vec{V}^{ac}\cdot\vec{V}^{ac*}$.

Even though $\mathcal{G}^{d-d+}$ and $\mathcal{G}^{b+b-}$ show a ground-state polarization (are nonzero), it is that of a CDW rather than an SDW, so diagram 2(d) gives an isotropic result. By these considerations the uniaxial anisotropy comes from diagrams 2(e)–2(h), with 2(e)–2(f) and 2(g)–2(h) being complex-conjugate pairs, so Ω is real. Here the $\vec{V}^{ij}$ of Eq. (9) are taken to be real, only 2(e) and 2(g) are evaluated, and each is given a factor of 2. When diagrams 2(e) and 2(g) are evaluated, using again the approximation that $g, h, kT \ll E_g$, the result can be manipulated using time-reversal symmetry[9] into the form

$$\Omega_{so} = \frac{4gh}{E_g^2 kT} \sum_{\vec{k}\nu} f(\vec{k}, \omega_\nu) \, [(\hat{n}\cdot\vec{V}^{bd+})(\hat{n}\cdot\vec{V}^{ca}) + (\hat{n}\cdot\vec{V}^{bd-})(\hat{n}\cdot\vec{V}^{ca})], \tag{10}$$

where $f(\vec{k}, \omega_\nu)$ also appears in the gap equation for g which is $1 = -(v_1/kT)\sum f(\vec{k}, \omega_\nu)$. Note that Ω_{so} is proportional to h and would be zero if the excited-state spin polarization were not included. By examining the symmetry properties of Ω_{so} it is straightforward to show that for an incommensurate $\vec{Q}$ in an otherwise cubic crystal Ω_{so} has uniaxial symmetry with respect to the $\vec{Q}$ direction and is therefore proportional to $(\hat{n}\cdot\hat{Q})^2$. Therefore, the gap equation for g can be used to define an average and obtain a compact result that displays the quantities determining the magnitude of Ω_{so} and the temperature dependence. Using also Eq. (7) to eliminate h, the result is

$$\Omega_{so} = -\frac{4g^2(T)v_2}{E_g^2 v_1^2} \langle V^{ca}(V^{db+} + V^{db-})\rangle (\hat{n}\cdot\hat{Q})^2 . \tag{11}$$

As mentioned in Sec. I, the temperature dependence is that of the gap, $g(T)$, squared.

The size of the anisotropy Ω_{so} can be estimated from Eq. (11) if the values of the various parameters are known. For chromium crude estimates can be made. The optical and magnetic properties imply $g(0) \cong 10^{-13}$ erg and $v_1 \cong 10^{-35}$ erg cm. The Coulomb matrix element v_2 involves more orbitals that are not identical than does v_1 and is therefore expected to be smaller, perhaps $v_2 \cong 10^{-1} v_1$. The atomic spin-orbit coupling constant for chromium, and band-structure calculations lead to a rough estimate that $V/E_g \cong 0.03$. Putting these values into Eq. (11) gives $\Omega_{so}(0) \cong 7.2 \times 10^5$ erg/cm^3, which is somewhat more than an order of magnitude greater than the value deduced from the phenomenological analysis. This will be discussed further in Sec. III.

The change in the free energy due to $\mathcal{H}_{dd}$ of Eq. (5), denoted Ω_{dd}, is also dealt with perturbatively. Because $\mathcal{H}_{dd}$ is a two-particle operator in spin and space coordinates, a uniaxial anisotropy arises in first order, and from matrix elements of the nesting states alone. The excited states need not be considered. Considerations like those employed in the discussion of $\mathcal{H}_{so}$ serve to eliminate all first-order diagrams but those of Fig. 3. Two equal but distinct diagrams are shown for 3(a), and the diagrams of 3(b), 3(c), and 3(d) also have equal partners, not shown. Again using the gap equation for g to define an average the contribution of diagram 3(a) is evaluated to be

$$\Omega_a = 4(g/v_1)^2 \langle \hat{n} \cdot \langle a\vec{k}, b\vec{k}+\vec{Q} | \overleftrightarrow{D}(r-r') | \times b\vec{k}+\vec{Q}, a\vec{k}' \rangle \cdot \hat{n} \rangle_{av} . \tag{12}$$

As in the case of spin-orbit coupling, the matrix element of $\overleftrightarrow{D}$ will reflect the uniaxial symmetry of the antiferromagnetic band structure for $\vec{Q}$ incommensurate. Taking the bands to be plane waves provides a poor approximation for the magnitude of the matrix element, but reveals the $\vec{Q}$ dependence clearly. Noting that $\overleftrightarrow{D}(\vec{r})$ can be written

$$\overleftrightarrow{D}(r) = (e^2\hbar^2/4m^2c^2)\vec{\nabla}\vec{\nabla}(1/|\vec{r}|) , \tag{13}$$

the matrix element of Eq. (13) in the plane-wave approximation is

$$\langle a\vec{k}, b\vec{k}'+\vec{Q} | \overleftrightarrow{D} | b\vec{k}+Q, ak' \rangle = (e\hbar^2/4m^2c^2) D(\vec{Q})\vec{Q}\vec{Q} , \tag{14}$$

where, per unit volume,

$$D(\vec{q}) = \int d^3r\, e^{-i\vec{q}\cdot\vec{r}} \frac{1}{|\vec{r}|} = \frac{4\pi}{|\vec{q}|^2} . \tag{15}$$

With this approximation, the contribution of diagram 3(b) and its partner is the same as from 3(a), and

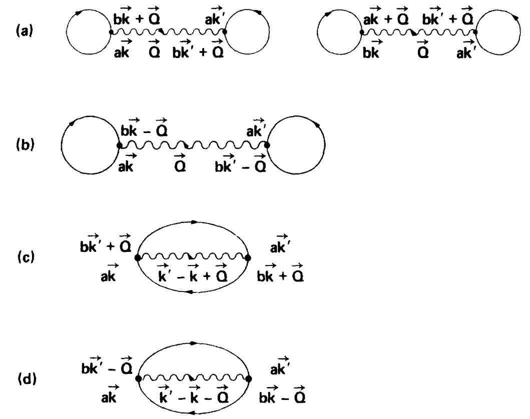

FIG. 3. Free-energy perturbation diagrams due to magnetic dipole-dipole interaction.

$$\Omega_a + \Omega_b = (g/v_1)^2 D_1 (\hat{n} \cdot \hat{Q})^2 , \tag{16}$$

where

$$D_1 \equiv 8\pi e^2\hbar^2/m^2c^2 . \tag{17}$$

Similarly the contributions from diagrams 3(c) and 3(d) can be evaluated, and Ω_{dd} is obtained as

$$\Omega_{dd} = (g/v_1)^2 (D_1 - D_2)(\hat{n} \cdot \hat{Q})^2 , \tag{18}$$

where D_2 is an average involving the gap equation of the type appearing in Eq. (12),

$$D_2 = (8\pi e^2\hbar^2 |\vec{Q}|^2/m^2c^2) \langle D(\vec{k}' - \vec{k} + \vec{Q}) \rangle_{av} , \tag{19}$$

and some isotropic terms independent of $\hat{n}$ have been dropped.

The main points to be noted about the result in Eq. (18) are the uniaxial symmetry and that the temperature dependence, like that of the spin-orbit result, is that of the gap squared. Also the order of magnitude can be estimated using values given above for g and v_1, and noting that $D_1 = 0.85 \times 10^{-38}$, to be $\Omega_{dd} \cong 8.5 \times 10^5$ erg/cm^3. This value is comparable to that of Ω_{so}, but could be much smaller if substantial cancellation between D_1 and D_2 occurs.

III. DISCUSSION

The calculation of Sec. II shows that the spin-dependent interactions usually associated with magnetic anisotropy do indeed result in a uniaxial anisotropy with respect to the $\vec{Q}$ direction for an incommensurate SDW. However, the temperature dependence of the anisotropy due to both mechanisms is the same, that of $g(T)^2$. Thus, it may be possible to obtain the correct polarization at low temperatures, but a spin flip will not occur because $g^2(T)$ goes monotonically to zero as T increases to T_N.

The passage of an anisotropy constant through zero in other materials has usually been accounted for by a competition between two sources of anisotropy that have similar magnitude, opposite sign, and a different temperature dependence. One example is competition between Ω_{dd} and Ω_{so} to explain the Morin transition in Fe_2O_3.[10] In this localized-spin case it is found that the two mechanisms have a different T dependence, in sharp and unfortunate contrast to the results obtained here for an itinerant (spin-$\frac{1}{2}$) system. The reason for this difference can be traced to the fact that for spin greater than $\frac{1}{2}$ it is not necessary to account for the spin polarization of the excited states [recall the dependence on h in Eq. (10)] to obtain a magnetic anisotropy from the spin-orbit interaction. There is a single-ion uniaxial anisotropy simply proportional to S_z^2, where S_z is a single-site spin operator, and the mean-field temperature dependence is that of $\langle S_z^2\rangle$ rather than the $\langle S_z\rangle^2$ obtained for the two-site dipole-dipole interaction,[10] and also obtained for both mechanisms in the itinerant (spin-$\frac{1}{2}$) case discussed here. For the itinerant system there does not appear to be any simple single-particle analog to the single-ion anisotropy of the localized system, because, as pointed out long ago by Van Vleck,[11] the algebra of the Pauli spin operators is such that powers of them can always be reduced to a linear form and this is not allowed in the anisotropy energy, which must involve even powers of the spin operators due to time reversal symmetry. Therefore, as mentioned in Sec. I, the model of the magnetic state, itinerant in the case at hand, influences qualitatively the result of the anisotropy calculation. The single-particle spin-orbit interaction must be combined with a two-particle interaction, leading to a temperature dependence like that of the two-particle dipole-dipole interaction in the HF approximation.

A second example is competition between the anisotropy due to orbitally degenerate states at different parts of the Fermi surface in an itinerant ferromagnet.[2] It is difficult to see how this second situation could apply directly for chromium for two reasons. First, the paired states producing antiferromagnetic gaps are orbital singlets, according to band calculations. Second, the pairing actually destroys a portion of the Fermi surface by opening up gaps, so that the paired states do not lie on the Fermi surface.

One possibility which is excluded by the present calculation lies in the observation that states which are not strongly paired are nonetheless spin polarized and contribute to the magnetic moment. While the energies of these states are not much altered by the SDW, they influence the energies of the strongly paired states by their contribution to the internal field responsible for pairing.[8,12,13] Some of these states will have orbital degeneracy and may lie on the Fermi surface. A numerical treatment of the effect of spin-orbit coupling on a realistic band structure might find a temperature dependence other than that obtained for the idealized band structure of Sec. II.

Another possibility is that there is some other microscopic mechanism not discussed here which produces an anisotropy of suitable size and a different temperature dependence. Such a mechanism could compete with the ones discussed here to produce the T dependence of Eq. (2). The fact that Ω_{so} and Ω_{dd} are estimated to be larger than the observed anisotropy would be very useful in a model based on competition if the two temperature dependences are very similar. (In Fe_2O_3 the two mechanisms must cancel within 2% to obtain the correct T dependence.[10])

In summary, the work described here is the first effort to give a microscopic theory of magnetic anisotropy for an incommensurate SDW material like chromium. A uniaxial anisotropy with respect to the Q direction has been demonstrated, and estimated to be larger than that observed in chromium. The method of the calculation could be readily extended to obtain a cubic anisotropy. The temperature dependence of the anisotropy is that of the antiferromagnetic gap squared, and therefore the spin-flip transition in chromium is not accounted for. If the basic itinerant model of the magnetic state is to be kept, future efforts must be directed towards examining the effects of a realistic band structure, or searching for an additional microscopic anisotropy mechanism.

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