Magnetoelastic coupling in helimagnets: A molecular-field theory of the $\frac{1}{4}$ lock-in phase of holmium

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A free energy formulated within the molecular-field treatment of a Heisenberg-like Hamiltonian serves as the basis for a model of the recently reported wave-vector lock-in at $\mathbf{Q}=\frac{1}{4}c^*$ in the incommensurate hexagonal helimagnet Ho. Variation of the exchange integral with interionic distance is shown to provide a mechanism for the observed decrease of Q as the temperature is lowered below $T_N \cong 131$ K. Umklapp terms of the form $\Delta_{4Q,G}$ are then responsible for stabilizing the $\frac{1}{4}$ commensurate phase over a narrow temperature region around 96 K. A crucial ingredient of the model is a symmetry-breaking term, which distorts the polarization of the spin density from helical to elliptical. Such an interaction can arise from the basal-plane component of an applied magnetic field or of uniaxial stress. Agreement between the model and experimental results for a wide range of magnetic properties is demonstrated using numerical estimates for a relatively small number of exchange parameters.

I. INTRODUCTION

Although the magnetic properties of heavy rare-earth metals have been the subject of intense research for more than 30 years, [1,2] interesting phenomena associated with these hexagonal helimagnets continue to be discovered. Recent attention has focused mainly on the magnetic ordering process of holmium and has revealed surprisingly complex low-temperature spin structures[3–6] and magnetic-field effects.[7–10] This work is an attempt to further understand the behavior of the magnetic wave vector $\mathbf{Q}(\|\hat{c})$, which characterizes the periodicity of the helical spin structure, at temperatures not too far below the Néel temperature ($T_N \cong 131$ K). In this regime, Q is incommensurable with the lattice periodicity and decreases as the temperature is lowered.[11] Of particular interest is the recent observation,[12,13] in a magnetic field applied along the c axis, of a wave vector lockin to a commensurate value $\mathbf{Q}=\frac{1}{4}\boldsymbol{c}^*$ over a narrow temperature range of a few degrees Kelvin near 96 K. It is shown here that a free energy derived within the molecular-field approximation, which includes magnetoelastic coupling through the variation of Heisenberg exchange interactions with interionic distance, can account for the temperature dependence of Q and that umklapp terms of the form $\Delta_{4Q,G}$ are then responsible for stabilizing the $\frac{1}{4}$ commensurate phase. An essential requirement of the model is a mechanism that distorts the polarization of the spin density from helical to elliptical. The in-plane component of an applied magnetic field or of uniaxial stress can provide the necessary symmetry-breaking term. A brief account of a completely phenomenological version of the present theory has been published[14] (referred to as I).

Apart from the early work of Elliott and Wedgwood[15] based on the Ruderman-Kittel-Kasuya-Yosida (RKKY) model (and low-temperature treatments of single-chain models[16]), theoretical investigation of the temperature dependence associated with helimagnet periodicities has received little attention. The model discussed in this work was inspired by the early observation of large magnetoelastic coupling in the heavy rare-earth metals[2,17] and recent data[13] on Ho showing lattice distortions associated with the lock-in, as well as by the exchange magnetostriction model of Lee.[18] Magnetic-coupling-induced relaxation of the lattice is known to give rise to nonlocal biquadratic exchange contributions to the usual Heisenberg Hamiltonian.[19] Within the context of a Landau-type expansion of the free energy derived within molecular-field theory, this relaxation introduces a nonlocal fourth-order term. As discussed in I, such nonlocality leads to a temperature dependence of the wave vector $\mathbf{Q}$.

Lockin phenomena have been studied for a wide variety of systems,[20,21] including helically modulated spin structures.[22–24] Commensurability effects in Ho due to magnetoelastic coupling, which differ from those investigated here, have been discussed by Vigren.[25] A consequence of expressing the Landau-type free energy as a function of Fourier components of the spin density, $\mathbf{S}_Q$, is the appearance of umklapp terms of the form[22,26] $(\mathbf{S}_Q\cdot\mathbf{S}_Q)^2\Delta_{4Q,G}$. Low-order terms (in $\mathbf{S}_Q$) determine the magnetic ordering process at temperatures not too far below T_N so that a commensurate phase, if it occurs, can be expected with periodicity *four*. In the case of hcp lattice structures (such as Ho), close-neighbor exchange interactions stabilize a (frustration-induced) incommensurability.[1,2] It is accidental in Ho that $Q(T)$ passes through a value $\frac{1}{4}c^*$, allowing for the possibility of umklapp-induced lock-in. Note that helical polarization may be described by taking[26] $\mathbf{S}_Q=|S_Q|(\hat{\mathbf{x}}+i\hat{\mathbf{y}})/\sqrt{2}$, in which case the umklapp terms are zero. It is for this reason that a distorting mechanism is essential to the present theory. A similar conclusion has been made by Harris, Rastell, and Tassi[24] in their analysis of lock-in effects associated with helimagnets at low temperatures.

The present theory demonstrates that the $\frac{1}{4}$ lock-in observed in Ho can be explained qualitatively by magnetoe-

lastic coupling. This is illustrated by a reasonably successful attempt to achieve agreement with experimental data based on a low-order expansion of the free energy and with only close-neighbor exchange interactions included. The lock-in temperature is sufficiently far below T_N, however, and the exchange interactions involving higher neighbors sufficiently large,[2,27] that these approximations may be expected to limit any quantitative agreement. In addition, although it can be argued that a helical-symmetry-breaking mechanism likely exists, the observed lock-in over a temperature range of 2 K can be explained by the present model only if this term in the free energy is rather large.

The remainder of this paper is organized as follows. The molecular-field theory of a free energy expanded to sixth order in the spin density, as well as an exchange magnetostriction model of fourth-order nonlocality, are presented in Sec. II. A parametrization of the spin density appropriate for hcp holmium is discussed in Sec. III. The temperature dependence of Q in the incommensurate phase is described in Sec. IV and numerical estimates made of low-order exchange and magnetoelastic coupling interactions which correlate a wide variety of thermodynamic data. Helical-symmetry breaking is introduced in Sec. V, demonstrating that the theory of umklapp-induced $\frac{1}{4}$ lock-in is in fair agreement with experimental data. A discussion of these results and some conclusions are made in Sec. VI.

II. MOLECULAR-FIELD THEORY AND MAGNETOELASTIC COUPLING

The starting point of the present model is the well-tested[2] assumption of localized spins interacting through Heisenberg exchange

$$\mathcal{H}=\tfrac{1}{2}\sum_{ij} J_{ij}\mathcal{s}_i\cdot\mathcal{s}_j \ , \tag{1}$$

where the configuration $\mathcal{s}\perp\hat{c}$ is stabilized by strong planar anisotropy[2] and $J_{ij}>0$ implies antiferromagnetic coupling. The method for obtaining a Landau-type expansion of the free energy derived within molecular-field theory given by Bak and von Boehm[28] is straightforward to generalize for the present case (also see Ref. 29). The result to sixth order in the spin density is

$$\begin{aligned} F_s=&(1/2V^2)\int d\mathbf{r}\int d\mathbf{r}'\,A(\tau)\mathbf{s}(\mathbf{r})\cdot\mathbf{s}(\mathbf{r}') \\ &+(B/4V)\int d\mathbf{r}[\mathbf{s}(\mathbf{r})\cdot\mathbf{s}(\mathbf{r})]^2 \\ &+(C/6V)\int d\mathbf{r}[\mathbf{s}(\mathbf{r})\cdot\mathbf{s}(\mathbf{r})]^3+\cdots \ , \end{aligned} \tag{2}$$

where $\mathbf{s}=\langle\mathcal{s}\rangle$ characterizes long-range order, $\tau=\mathbf{r}'-\mathbf{r}$, and

$$A(\mathbf{r})=ak_BT\delta(\mathbf{r})+j^2J(\mathbf{r}) \ , \tag{3}$$

$$B=bk_BT, \quad C=ck_BT, \ldots . \tag{4}$$

The coefficients $a,b,c,\ldots$ depend on (in the case of the heavy rare-earth metals) the *total* angular momentum j, through the Brillouin function, and may be expressed by

$$a=3j/(j+1), \quad b=\tilde{b}a^4, \quad c=(3\tilde{b}^2a-\tilde{c})a^6 \ , \tag{5}$$

$$\tilde{b}=\{[(2j+1)^4-1]/(2j)^4\}/45 \ , \tag{6}$$

$$\tilde{c}=2\{[(2j+1)^6-1]/(2j)^6\}/945 \ , \tag{7}$$

with $j=8$ for Ho. Note that only the second-order term in the free energy (2) is nonlocal. Also note that the Hamiltonian (1) omits anisotropy *within* the hexagonal plane,[2] which would make a contribution at sixth-order in $\mathbf{s}$. The possibility that such an interaction could effect the lock-in process discussed below was investigated and found not to be relevant at the (high) temperatures of interest here.

Magnetoelastic coupling is accounted for within the present model by allowing for variation of the exchange interaction with interionic distance.[18] This effect is described by writing $\mathbf{r}=\mathbf{r}_0+\mathbf{u}(\mathbf{r}_0)$ (where $\mathbf{r}_0$ characterizes equilibrium positions in the paramagnetic state), so that to low order

$$J(\mathbf{r}'-\mathbf{r})=J(\mathbf{r}_0'-\mathbf{r}_0)+[\mathbf{u}(\mathbf{r}_0')-\mathbf{u}(\mathbf{r}_0)]\cdot\nabla J(\mathbf{r}_0)+\cdots \ . \tag{8}$$

With the expansion

$$\mathbf{u}(\mathbf{r}')=\mathbf{u}(\mathbf{r})+\tau_\alpha\frac{\partial\mathbf{u}}{\partial r_\alpha}+\cdots \ , \tag{9}$$

where $\alpha=x,y,z$ and the summation convention is used, relation (8) can be expressed as

$$J(\tau)\cong J(\tau_0)+e_iK_i(\tau_0) \ , \tag{10}$$

where $i=1-6$ in the Voigt notation, e_i is the strain tensor, and the symmetric magnetoelastic coupling coefficients are given by

$$K_{\alpha\beta}(\tau_0)=\tfrac{1}{2}\left[\frac{\partial J}{\partial r_\alpha}\tau_\beta+\frac{\partial J}{\partial r_\beta}\tau_\alpha\right]_0 \ . \tag{11}$$

With the addition of the elastic energy, strain contributions to the free energy are thus given by (also see Ref. 26)

$$F_e=(\tfrac{1}{2}j^2/V^2)\int d\mathbf{r}\int d\mathbf{r}'K_i(\tau)e_i\mathbf{s}(\mathbf{r})\cdot\mathbf{s}(\mathbf{r}')+\tfrac{1}{2}\omega C_{ij}e_ie_j \ , \tag{12}$$

where $\omega=V/N$ is the unit-cell volume and the subscript 0 on position variables has been omitted for convenience. The strain tensor at equilibrium is determined by $\partial F_e/\partial e_i=0$ so that

$$e_i=(-\tfrac{1}{2}j^2/\omega V^2)\int d\mathbf{r}\int d\mathbf{r}'\mathrm{s}_{ij}K_j(\tau)\mathbf{s}(\mathbf{r})\cdot\mathbf{s}(\mathbf{r}') \ , \tag{13}$$

where s_{ij} is the compliance tensor, $\mathrm{s}=\mathrm{C}^{-1}$. Inserting this expression back into (12) demonstrates that the effect of magnetoelastic coupling is a *nonlocal* contribution to $F[\mathbf{s}(\mathbf{r})]$ at fourth order (also see Ref. 30):

$$F_K=(-\tfrac{1}{8}j^4/\omega V^4)\int d\mathbf{r}_1\int d\mathbf{r}_2\int d\mathbf{r}_3\int d\mathbf{r}_4K_i(\mathbf{r}_1-\mathbf{r}_2)\mathrm{s}_{ij}K_j(\mathbf{r}_3-\mathbf{r}_4)[\mathbf{s}(\mathbf{r}_1)\cdot\mathbf{s}(\mathbf{r}_2)][\mathbf{s}(\mathbf{r}_3)\cdot\mathbf{s}(\mathbf{r}_4)] \ . \tag{14}$$

This term is to be added to the free energy (2) with the understanding that $\mathbf{r}$ denotes equilibrium positions with respect to the paramagnetic phase, $F=F_s\{\mathbf{r}_0\}+F_K\{\mathbf{r}_0\}$. Note that the structure of (14) differs from the type of nonlocality assumed in I.

III. APPLICATION TO hcp HOLMIUM

Following the notation in I, the hcp crystal structure of Ho ($P6_3/mmc$) can be described by a simple hexagonal lattice with a two-site basis labeled A and B with vectors $\mathbf{w}_A=\mathbf{0}$ and $\mathbf{w}_B\equiv\mathbf{w}=\frac{1}{2}a\hat{\mathbf{x}}+\frac{1}{3}\mathscr{A}\hat{\mathbf{y}}+\frac{1}{2}c\hat{\mathbf{z}}$, where a and c are the lattice constants, $\mathscr{A}=\sqrt{3}a/2$, and $\hat{\mathbf{x}}\perp\hat{\mathbf{y}}\perp\hat{\mathbf{z}}$. The spin density is assumed to have the form[26]

$$\mathbf{s}(\mathbf{r})=(V/2N)\sum_{\mathbf{R}_1}\sum_j\boldsymbol{\rho}_j(\mathbf{r})\delta(\mathbf{r}-\mathbf{R}) \tag{15}$$

with $j=A,B$ and $\mathbf{R}=\mathbf{R}_1+\mathbf{w}_j$, where $\mathbf{R}_1$ denotes hexagonal Bravais lattice sites, and with long-range order characterized by

$$\boldsymbol{\rho}_j(\mathbf{r})=\mathbf{S}_j e^{i\mathbf{Q}\cdot\mathbf{r}}+\mathbf{S}_j^* e^{-i\mathbf{Q}\cdot\mathbf{r}} . \tag{16}$$

As discussed in I, a sufficiently flexible parametrization of the polarization vectors is expressed by

$$\mathbf{S}_A=S(\cos\beta\hat{\mathbf{x}}+i\sin\beta\hat{\mathbf{y}})e^{i\gamma} \tag{17}$$

and $\mathbf{S}_B=\mathbf{S}_A e^{i\gamma'}$, allowing for helical ($\beta=\pi/4$), linear ($\beta=0$ and $\beta=\pi/2$), and elliptical ($0<\beta<\pi/2$) polarizations of the magnetic order. With this description of the spin density, the free energy (2) simplifies to

$$F_s=\tfrac{1}{2}aTS^2+\tfrac{1}{2}j^2(J_Q+\tilde{J}_Q\cos\phi)S^2+(B/16)S^4[2+\cos^2(2\beta)]+(C/16)S^6[\tfrac{2}{3}+\cos^2(2\beta)]$$
$$-(1/16)S^4[(B+CS^2)\cos^2(2\beta)\sin(2\phi)\sin(2\psi)]\Delta_{4Q,G} , \tag{18}$$

where $\phi=\gamma'-\gamma$, $\psi=\phi+\phi'$, and

$$\tilde{J}_Q=(1/N)\sum_{\mathbf{R}_1}J(\mathbf{R}+\mathbf{w})e^{i\mathbf{Q}\cdot(\mathbf{R}_1+\mathbf{w})} , \tag{19}$$

with J_Q similarly defined but with $\mathbf{w}=\mathbf{0}$. Note that the umklapp term in (18) is zero for helical polarization and also if the sublattice phase angle difference ϕ is zero.

For hexagonal symmetry, only two independent magnetoelastic coupling constants occur,[26] $K_{xx}=K_{yy}$ and K_{zz}. Numerical estimates of these two parameters based on the stress dependence of the Néel temperature[31] of Ho (also see below) reveals, however, that contributions to the free energy from K_{zz} are much larger than those from K_{xx}. The nonlocal terms (14) are thus well approximated here by

$$F_K\cong-\tfrac{1}{8}j^4(s_{33}/\mathscr{A})[K_Q+\tilde{K}_Q\cos\phi]^2S^4 , \tag{20}$$

where $K\equiv K_{zz}$ and with $\tilde{K}_Q$ and K_Q defined as by (19). Uniform strain along the c axis from (13) is determined by

$$e_3\cong-\tfrac{1}{2}j^2(s_{33}/\mathscr{A})[K_Q+\tilde{K}_Q\cos\phi]S^2 . \tag{21}$$

Ferromagnetic exchange interactions in the basal plane stabilize a configuration $\mathbf{Q}\|\hat{\boldsymbol{c}}$ so that the wave-vector dependence is characterized by the interlayer turn angle $q=\frac{1}{2}Qc$. It is then convenient to follow Stringfellow *et al.*[32] and write

$$J_Q=(2/N)\sum_{n=0}J_{2n}\cos(2nq) , \tag{22}$$

$$\tilde{J}_Q=(2/N)\sum_{n=0}J_{2n+1}\cos[(2n+1)q] , \tag{23}$$

where J_n represents all coupling between magnetic ions separated by n layers. For simplicity, only the lowest-order interactions $J_0\cong3J(\boldsymbol{a})$, $J_1\cong3J(\mathbf{w})$, and $J_2\cong J(\boldsymbol{c})$ are included in the following analysis. K_Q and $\tilde{K}_Q$ similarly defined, with $K_0=0$, $K_1\cong\frac{3}{2}c\partial J(\mathbf{w})/\partial z$, and $K_2\cong c\partial J(\boldsymbol{c})/\partial z$.

IV. INCOMMENSURATE PHASE

Analysis of the free energy for the incommensurate (IC) phase demonstrates the magnetoelastic-induced temperature dependence of Q and allows for estimates to be made of low-order exchange and magnetostriction coupling constants by comparison with a variety of experimental data. In this case F, given by (18) plus (20), is minimized with a helical polarization $\beta=\pi/4$ and a sublattice phase angle difference $\phi=0$, so that to fourth order in S

$$F_{IC}\cong\tfrac{1}{2}aTS^2+\tfrac{1}{2}j^2(J_Q+\tilde{J}_Q)S^2+\tfrac{1}{8}BS^4$$
$$-\tfrac{1}{8}j^4(s_{33}/v)(K_Q+\tilde{K}_Q)^2S^4 . \tag{24}$$

With only close-neighbor coupling included (as described above), the equilibrium condition $\partial F/\partial Q=0$ yields the relation

$$J_1+4J_2\cos q-j^2(s_{33}/\mathscr{A})$$
$$\times[K_1\cos q+K_2\cos(2q)][K_1+K_2\cos q]S^2=0 \tag{25}$$

where temperature dependence enters through $S^2\sim T_N-T$. Numerical results for $q(T)$ are presented in the next section. At values of T close to T_N, where S^2 is small, (25) simplifies to

$$\cos q\cong c_0+\tfrac{1}{4}j^2(s_{33}/\mathscr{A}J_2)$$
$$\times[K_1+4c_0K_2][c_0K_1-K_2+2c_0K_2]S^2 \tag{26}$$

where $c_0=-J_1/4J_2$. Using the estimated values of exchange parameters given below, this relation illustrates

that q decreases as the temperature is lowered. Note that the result $\cos q = c_0$ at T_N is well known.[2]

Estimates of the exchange parameters J_0, J_1, and J_2 were made as follows. For J_1, the value determined from the spin-wave data of Nicklow *et al.*[27] at 48 K is used (accounting for the factor of -2 difference between Hamiltonians). (It should be noted that there is a wide range of estimates for this parameter.[27,32]) The above expression for $\cos q$ at T_N, with[11] $q = 50°$, can then be used to estimate J_2. Finally, the Néel temperature for the present model is given by (see, e.g., Coqblin[1])

$$T_N = (j^2/a)[2J_2 - 2J_0 + J_1^2/(4J_2)] \tag{27}$$

so that an estimate for J_0 may then be made. This analysis yields the values $J_0 = -1.05$, $J_1 = -2.37$, and $J_2 = 0.922$ K. The in-plane order is ferromagnetic and, within the framework of this close-neighbor model, the helical modulation along the c axis is stabilized by the frustration characteristic of antiferromagnetic next-nearest-neighbor interactions.[2] As noted in the Introduction, higher-neighbor exchange interactions are not negligible[27] so that J_0 and J_2 as estimated above may be considered as effective coupling constants, which are renormalized by the longer-range interactions not explicitly included in the model. The Curie temperature,

$$T_0 = -2(j^2/a)(J_0 + J_1 + J_2) ,$$

calculated with these parameters is 60 K, in fair agreement with the experimental estimate[33] of 80 K.

Crude estimates of the magnetoelastic coupling parameters K_1 and K_2 were made based on a comparison of expression (21) for the c-axis strain and experimental data[34] for $\Delta c/c$ (Ref. 35), s_{33} (Ref. 36), q and S^2 (Ref. 11) at 95 K and 120 K. Note that neutron-scattering intensity is proportional to S^2 and that $S^2 = 2$ at $T = 0$. This analysis yields the estimates $K_1 = -23$ K and $K_2 = -30$ K. As a check on these values, they may be used to estimate the pressure dependence of the Néel temperature,[26] with the result $\partial T_N/\partial p \cong -0.33$ K/kbar. This value is in good agreement with the experimental estimates[37] -0.48, -0.33, and -0.40 K/kbar. Note that the term involving K_2 in the free energy (20) appears as $K_2 \cos(2q)$ so that in the temperature range of interest (where $q \cong \pi/4$), magnetoelastic effects are relatively insensitive to its value. In the analysis of the wave-vector lock-in that follows, the value of K_2 is adjusted to best reproduce the observed $q(T)$.

V. $\frac{1}{4}$ LOCK-IN

As emphasized in I and in the Introduction, the observed[12] wave-vector lock-in at $q = \pi/4$ is obtained within the present formalism only if a helical-symmetry-breaking term is added to the free energy. The simplest type of interaction, which achieves this effect, is of the form

$$F_p = A_p S^2 \cos^2\beta , \tag{28}$$

which could arise from an in-plane component of the uniform magnetization[38] $\mathbf{m}$, $A_p = \frac{1}{4}Bm_\perp^2$, or uniaxial stress[26] σ_i, $A_p = -j^2 k \sigma_\perp (s_{11} - s_{12})$, where k is a magnetoelastic coupling coefficient ($k \ll K$). For the analysis that follows, A_p is treated as a phenomenological parameter. The results presented in this section were obtained by direct numerical minimization of the free energy (18), (20), and (28), $F = F_s + F_K + F_p$, as a function of the variables S, β, ϕ, ... Due to the umklapp term in (18), the IC and commensurate (C) cases must be treated separately: $F_{IC} = F(S,\beta,\phi,\psi,q)$ and $F_c = F(S,\beta,\phi,\psi)$ with $q = \pi/4$. The relative stability of IC and C phases can then be checked by comparing F_{IC} and F_C as a function of temperature.

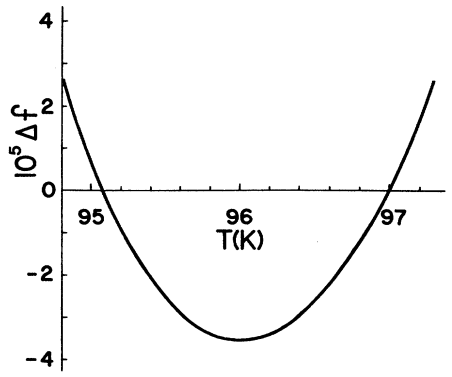

FIG. 1. Numerically determined relative free energy difference $f = (F_{IC} - F_C)/F_{IC}$ showing the stability of the commensurate phase for 95.04 K $\lesssim T \lesssim$ 97.00 K.

Computations were performed using the previously described values for J_0, J_1, J_2, and K_1. K_2 was treated as a fitting parameter so that $q(T^*) = \pi/4$ in the IC phase, where $T^* = 96$ K, and A_p was adjusted to reproduce the temperature width of the lock-in, about 2K. Reasonable results were obtained using the values $K_2 = -18.3$ and $A_p = 7$. Note that this value for K_2 gives a result for the pressure dependence of T_N, $\partial T_N/\partial p = -0.45$ K/kbar,

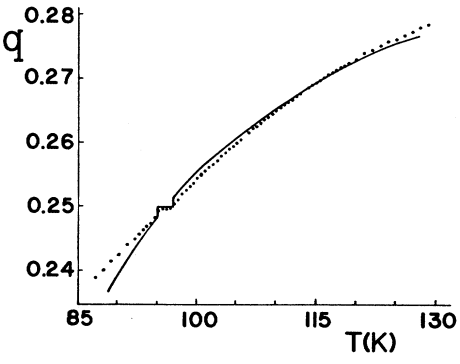

FIG. 2. Calculated results (solid curve) and experimental data of Ref. 12 (dotted curve) showing the temperature dependence of the interlayer turn angle $q = \frac{1}{2}Q$ and the lock-in at $q = \pi/4$.

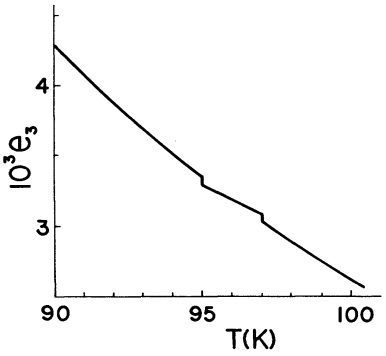

FIG. 3. Effects of the lock-in on the c-axis strain e_3.

within the range of experimental estimates. The relative free energy difference between the two phases shown in Fig. 1 demonstrates that $F_C < F_{IC}$ in a temperature interval $T_1 < T < T_2$, where $T_1 \cong 95.04$ K and $T_2 \cong 97.00$ K, and that the transitions at T_1 and T_2 are first order. Results for $q(T)$ shown in Fig. 2 illustrate further the stability of the $\frac{1}{4}$ commensurate phase and that the magnetoelastic coupling model can reasonably account for thermal effects on the interlayer turn angle of this helimagnet. The discrepancy at low temperatures is likely a consequence of retaining only low-order terms J_n and K_n as well as in the s^2 expansion of the free energy. Finally, the effects of the lock-in on the c-axis strain shown in Fig. 3 are consistent with the large anomalies recently reported by Tindall *et al.*[13] in $\partial e_3/\partial T$.

The principle effect of the symmetry-breaking term F_p is to distort the polarization of the spin density from helical to elliptical (in both IC and C phases), with $\beta \cong 0.27\pi$ at T near T^*. For T close to T_N, however, the result of this term to stabilize a linearly polarized state,[38] $\beta = \pi/2$ (for $A_p > 0$). The elliptical-linear transition is second order and in the present case occurs at $T_\beta \cong 124$ K. The possibility that such a transition takes place in Ho has already been proposed.[7,38] Also note that the phase angle sum ψ takes a value $\pi/4$ for both IC and C order, and that $\phi = 0$ in the IC phase, whereas $\phi \gtrsim 0$ in the C phase.

VI. DISCUSSION AND CONCLUSIONS

This work has demonstrated that magnetoelastic coupling due to the variation of the exchange integral with interionic separation leads to a temperature dependence of the incommensurate wave vector associated with helimagnets. The observed data for $q(T)$ of Ho is well accounted for by this model using a relatively small number of exchange and magnetoelastic coupling parameters, which correlate a wide variety of experimental results. A helical-symmetry breaking term is required, however, to account for the umklapp-assisted $\frac{1}{4}$ commensurate lock-in. A weak point of the theory is that the size of the coefficient A_p of this term required to reproduce the data of Ref. 12 is rather large. It can be shown using the expressions for A_p after (28) [and[26] $m_\perp \cong \chi H$, with $\chi^{-1} = a(T_N - T_0)$] that an in-plane magnetic field of over 1 T or uniaxial stress of over 10 kbar, for example, would be necessary to give a value $A_p = 7$ K. Such distortions are an order of magnitude larger than can be reasonably expected from the experimental setup of Ref. 12, where a magnetic field of 2.2 T was applied along the c axis. Helical symmetry is not broken in the case of $\mathbf{H} \perp \mathbf{S}$. A source of this discrepancy may be the omission (for simplicity) of longer-range exchange and magnetoelastic coupling interactions in the model. In addition, the present model omits effects arising from magnetoelastic-coupling-induced inhomogeneous strain (see below). It is clear, however, that the present theory contains the essential ingredients to explain $\frac{1}{4}$ lock-in transitions in helimagnets.

Some additional support for the existence of a helical-symmetry breaking mechanism can be found in recent data[13] on Ho. Peaks in the neutron-scattering intensity were observed at $2\mathbf{Q}$, with associated lock-in at $2q = \pi/2$. These data were taken in a magnetic field of 3 T applied along the c axis. It has been shown by Walker[39] that an induced periodic lattice distortion characterized by a wavevector $2\mathbf{Q}$ is expected to be absent in the case of perfect helical polarization but that it should occur in association with elliptical or linear polarization of the spin density wave. In these latter cases, a harmonic of the spin density at $3\mathbf{Q}$ is also expected. Further experiments to determine more precisely the character of the magnetic order in Ho are desirable.

Note added. After the completion of this work, evidence for a devil's-staircase behavior in Ho was reported[40] to result from a magnetic field applied along the c axis. A series of commensurate wavevector values were observed at low temperatures, in a regime where a finite Landau-type expansion of the free energy is likely to be meaningless. There is no particular reason to suspect that such effects are caused by magnetoelastic coupling; rather, the model of Ref. 28 appears to be relevant.[40]

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ACKNOWLEDGMENTS

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