Magnetic-field-induced phase transitions in heavy rare earth metals

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Rare earth metals having a "ribbon" singularity on the Fermi surface and a pronounced single-ion anisotropy in an external magnetic field are analyzed. A ferromagnetic phase is produced above a certain critical field. At low temperatures, only the lower of the spin-wave branches which exist in this ferromagnetic phase has dispersion. The behavior of the magnetic-structure vector as a function of the field and the temperature is determined by the ratio of the gap in the electron spectrum to the energy separation of the chemical potential from the critical point in the electron spectrum. The Curie temperature cannot change by a factor greater than three as the anisotropy constants are varied from zero to infinity. The rare earth metal is transformed from the paramagnetic phase to a helicoidal rather than ferromagnetic phase if the ribbon singularity on the Fermi surface exceeds a certain critical size. The constants of the microscopic theory can be determined by measuring the critical field and the longitudinal and transverse magnetizations of the transition from a ferromagnetic helix to a ferromagnet. At this transition the longitudinal magnetization will change linearly, $\sim(h_c - h)$, while the transverse magnetization will vary as the square root, $\sim(h_c - h)^{1/2}$.

1. INTRODUCTION

A broad spectrum of helicoidal structures is known to exist in heavy rare earth metals.[1] These structures have been attributed[2,3] to singular regions on the Fermi surface; the wave vector Q which "measures" the extremal cross section of this region determines the period $\sim 2\pi/Q$ and the direction of the corresponding helix. The magnetic anisotropy in a rare earth metal is produced primarily by the crystal field[1,4] and determines to a large extent both the diversity of long-period structures and their conversions into each other.[1]

The standard method[4] for describing effects of a single-ion anisotropy in a rare earth metal is based on the approach suggested by Callen and Callen.[5] This approach is used extensively to analyze experiments on rare earth metals and their compounds.[3,4,6] As we will show in the Appendix, however, this approach is not appropriate for describing rare earth metals. This fact was pointed out in Refs. 7–9 and a "mean-field anisotropy approximation" was proposed for seeking the ground state, the functional dependence of the magnetization on the external field, and the thermodynamics. The idea of this approximation is to discard the correlations and to minimize the free energy with respect to both the magnitude of the magnetization and the angles between the magnetic moment and the crystallographic axes. In order to use this method we need to know the particular region of parameters in which we can introduce $\langle J^x \rangle \neq 0$, $\langle J^y \rangle \neq 0$ (Ref. 10). For example, Zaĭtsev[11] showed in the Heisenberg model that for the case $S = 1$ with an anisotropy $\Sigma D (S_f^z)^2$ with $D > 0$ there is a region of the parameters of the system in which a state with a zero spin projection and with a quantization axis along the z axis is stable (in the classical description,[1] the moment "lies" in the plane perpendicular to the z axis in the same parameter region).

The stability of the ground state of a magnetic material and its low-temperature thermodynamics are determined by the spectrum of spin waves. This spectrum in rare earth magnetic materials has been the subject of several studies.[3,6,12] The standard approach is based on the use of the Holstein-Primakoff representation (see the bibliography in the book by Taylor and Darby[3]). As will be seen below, for the normal arrangement of the levels of an f ion this calculation method can be used to find the lower branch of the spin-wave spectrum, but it breaks down even at temperatures on the order of the distance between the lower levels of the f ion in the crystal field and a magnetic field. Vedyaev and Nikolaev showed,[6] without allowance for magnetic anisotropy, that singular sections (of the ribbon type, for example) on the Fermi surface give rise to a maximum in the electron susceptibility $\chi(\mathbf{q},0)$ at $\mathbf{q} = \mathbf{Q}$. This maximum corresponds to the extremal size of the singular region. Correspondingly, it corresponds to a minimum in the spin-wave spectrum at the same $\mathbf{Q}$. Vedyaev and Nikolaev[6] also gave equations for finding the spin-wave spectrum in an easy-plane, ferromagnetically ordered rare earth metal. Vedyaev and Nikolaev[6] took the single-ion anisotropy into account by introducing a phenomenological constant $B(T)$ in the gap of the spin-wave spectrum; working from the study by Callen and Callen,[5] they concluded that this contribution to the gap depends on the temperature as the tenth power of the magnetization. As already mentioned, this assertion is incorrect in the case of rare earth metals. We will show below that Eq. (7) of Ref. 6 is nevertheless valid for describing the lower branch of the spin-wave spectrum—at low temperatures, but for an easy-axis rather than easy-plane magnetic material. A method for correctly dealing with the single-ion anisotropy was pointed out in Refs. 11 and 13, and it was applied to rare earth metals in Ref. 14. What amounts to a general recipe for using the method was given in Ref. 11, although the illustrative calculations there dealt exclusively with the case $S = 1$ and $H_{\text{an}} = \Sigma D (S_f^z)^2$ for dielectrics. The same approach was used

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in Ref. 14 to derive an equation for the spin-wave spectrum in easy-axis hexagonal crystals of rare earth metals for arbitrary J in an *s-f*-exchange model.

In this paper we analyze the anomalous features of the low-temperature dependence of the magnetization of easy-axis rare earth metals on an external magnetic field. Our analysis is based on spin-wave theory; the single-ion anisotropy is taken into account exactly. The calculations yield for the magnetization simple expressions which can be used to find the constants of the microscopic theory from experiment. We also derive equations for plotting the (T,h) phase diagram of the magnetic states of rare earth metals.

2. HAMILTONIAN AND EQUATION FOR THE SPIN-WAVE SPECTRUM

We consider a system of localized *f* electrons and collectivized *c* electrons which are interacting with each other. Each *f* ion has its own term by virtue of the pronounced intraatomic and *LS* interactions. Each such ion is furthermore in a hexagonal crystal field. Using the leading term in the *s-f* exchange Hamiltonian for a rare earth metal,[15,16] and adding the energy of the single-ion magnetic anisotropy, we find the following Hamiltonian for the model:

$$H=H_0^c+H_0^f+H_i^{c-f},$$

$$H_0^c=\sum_{\mathbf{k}\sigma}\tilde{\xi}_{\mathbf{k}}^{\sigma}c_{\mathbf{k}\sigma}^{+}c_{\mathbf{k}\sigma},\qquad \tilde{\xi}_{\mathbf{k}}^{\sigma}=\varepsilon_{\mathbf{k}}-\mu-\frac{1}{2}g_c h\eta(\sigma),$$

$$H_0^f=\sum_{\mathbf{f}}[-hg_fJ_{\mathbf{f}}^z-D_2^0(J_{\mathbf{f}}^z)^2-D_4^0(J_{\mathbf{f}}^z)^4-D_6^0(J_{\mathbf{f}}^z)^6], \tag{1}$$

$$H_i^{c-f}=-(A/N)\sum_{\mathbf{kqf}}\sum_{\alpha\beta}e^{-i\mathbf{qf}}\mathbf{J}_{\mathbf{f}}\boldsymbol{\sigma}_{\alpha\beta}c_{\mathbf{k}\alpha}^{+}c_{\mathbf{k}+\mathbf{q}\beta}.$$

The operator J is the total angular momentum of the ion; i.e., the *LS* coupling of the *f* electrons is taken into account even in the zeroth approximation. In addition, the D_{2n}^0 ($n = 1, 2, 3$) are the constants of the single-ion anisotropy which correspond to the hexagonal symmetry of the rare earth metal.

For the calculations it is convenient to adopt the representation of Hubbard operators $X^{pq} = |p\rangle\langle q|$, which obey the multiplication rules $X^{mn}X^{pq} = \delta_{np}X^{mq}$ at a common site, while at different sites they commute in this case. The angular momentum operators $\mathbf{J}$ can then be expressed in terms of the Hubbard operators X as follows:

$$J_{\mathbf{f}}^z=\sum_{m=-J}^{J}mX_{\mathbf{f}}^{mm},\qquad J_{\mathbf{f}}^{\pm}=\sum_{m=-J}^{J}J_{m\pm1,m}^{\pm}X_{\mathbf{f}}^{m\pm1,m},$$

$$J_{m\pm1,m}^{\pm}=[J(J+1)-m(m\pm1)]^{1/2};$$

the number m specifies the projection of the angular momentum $\mathbf{J}$ on the quantization axis. After we introduce the mean-field approximation in the standard way, Hamiltonian (6) becomes

$$H_0^f=\sum_{\mathbf{f}m}E_mX_{\mathbf{f}}^{mm},\qquad E_m=-\left(hg_fm+\sum_{n=1}^{3}D_{2n}^0m^{2n}+2AR_c^zm\right),$$

$$H_0^c=\sum_{\mathbf{k}\sigma}\xi_{\mathbf{k}}^{\sigma}c_{\mathbf{k}\sigma}^{+}c_{\mathbf{k}\sigma},\qquad \xi_{\mathbf{k}}^{\sigma}=\tilde{\xi}_{\mathbf{k}}^{\sigma}-\eta(\sigma)AR_f^z,$$

$$R_c^z=\frac{1}{2N}\sum_{\mathbf{k}}[n(\xi_{\mathbf{k}}^{\uparrow})-n(\xi_{\mathbf{k}}^{\downarrow})],$$

$$R_f^z=\sum_m mN_m,\qquad n(\xi)=[e^{\beta\xi}+1]^{-1},\qquad N_m=\langle X_{\mathbf{f}}^{mm}\rangle,$$

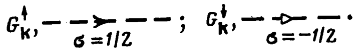
$$H_i^{c-f}=H_{i\parallel}^{c-f}+H_{i\perp}^{c-f},$$

$$H_{i\parallel}^{c-f}=-\sum_{\mathbf{fkq}m}e^{-i\mathbf{qf}}m\delta X_{\mathbf{f}}^{mm}(\overline{c_{\mathbf{k}\uparrow}^{+}c_{\mathbf{k}+\mathbf{q}\uparrow}}-\overline{c_{\mathbf{k}\downarrow}^{+}c_{\mathbf{k}+q\downarrow}}),$$

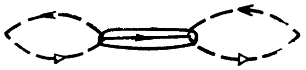
$$H_{i\perp}^{c-f}=-\frac{A}{N}\sum_{\mathbf{fkq}m}e^{-i\mathbf{qf}}(J_{m+1,m}^{+}X_{\mathbf{f}}^{m+1,m}c_{\mathbf{k}\downarrow}^{+}c_{\mathbf{k}+q\uparrow}$$

$$+J_{m-1,m}^{-}X_{\mathbf{f}}^{m-1,m}c_{\mathbf{k}\uparrow}^{+}c_{\mathbf{k}+q\downarrow}).$$

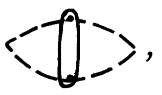
Here $c^+c = c^+c - \langle c^+c\rangle$, and $\delta X = X - \langle X\rangle$. The spin-wave spectrum can be found from the poles of the transverse spin correlation function. We seek this function by using a diagram technique for Hubbard operators[11,14] (this technique is similar to the spin technique of Ref. 17). We denote the Green's functions of the X operators by a solid line with an arrow, while the *c*-electron functions are represented by a dashed line with an arrow:

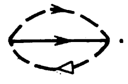


It does not matter just which ends—the *c*-electron or *f*-electron ends—we use to seek the correlation function, since the poles of these functions coincide with an interacting system. In zeroth order, the unknown function $\langle T\sigma^+(\tau)\sigma^-(\tau')\rangle$ is described by an ordinary electron loop. At low temperatures, diagrams of the type

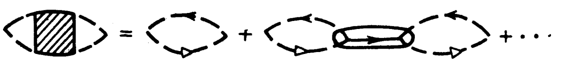


do not have any small factors on the order of Ag_0 (where $g_0\sim 1/2\mu$). Diagrams which have disconnected parts within an oval (diagram *a* of the two diagrams, just below) are exponentially small,[17] while diagrams which contain elements of type *b* introduce a small factor $\sim Ag_0\ln(\mu/A)$:


a


b

The unknown function is thus given by the series


(2)

Writing the sum of series (2) analytically, and making an analytic continuation to the upper half-plane, we find an equation for its poles[14]:

$$1+A^2\chi_0^{+-}(\mathbf{q},\omega)K_0(\omega)=0, \tag{3}$$

where

$$\chi_0^{+-}(\mathbf{q},\omega)=\sum_{k}\frac{n(\xi_{\mathbf{k}+\mathbf{q}}^{\downarrow})-n(\xi_{\mathbf{k}}^{\uparrow})}{\omega-(\xi_{\mathbf{k}+\mathbf{q}}^{\downarrow}-\xi_{\mathbf{k}}^{\uparrow})},$$

$$K_0(\omega)=\sum_m\frac{|J_{m+1,m}^{+}|^2\Delta N_{m+1,m}}{\omega-\Delta E_{m,m+1}},$$

$$\Delta N_{m+1,m}=N_{m+1}-N_m, \qquad \Delta E_{m,m+1}=E_m-E_{m+1}. \tag{4}$$

In the absence of an anisotropy, the spectrum of an f ion becomes equidistant; i.e., for each value of m we have $\Delta E_{m,m+1} = \Delta E$. From (2) and (3) we then find the standard result of the $s-f$ model,[17] since, as is easily shown, we have

$$\sum_m |J^{+}_{m+1,m}|^2 \Delta N_{m+1,m}=2\langle J^z\rangle.$$

The poles of the function $\chi_0{}^{+-}(\mathbf{q},\omega)$ lies in the region $\omega\sim\Delta_c = 2AR^{\,2}_{f}$ at small values of $\mathbf{q}$. Here we are interested in values $\omega\sim\omega_q\sim A^{\,2}g_0 \ll AR_f{}^z$, so we will be ignoring the dependence of the function $A^{\,2}\chi_0{}^{+-}(\mathbf{q},\omega)$ on the frequency ω. In this case, of course, the quantity $A^{\,2}\chi_0{}^{+-}(\mathbf{q},0)$ plays the role of a Fourier transform of the exchange integral between transverse components of the angular momentum, and Eq. (7) also describes spin waves in an anisotropic dielectric with a Hamiltonian

$$H=H_0{}^f-\sum_{\mathfrak{f}\mathfrak{f}'}\left[A_{\mathfrak{f}\mathfrak{f}'}S_{\mathfrak{f}}{}^zS_{\mathfrak{f}'}{}^z+B_{\mathfrak{f}\mathfrak{f}'}(S_{\mathfrak{f}}{}^+S_{\mathfrak{f}'}{}^-+S_{\mathfrak{f}}{}^-S_{\mathfrak{f}'}{}^+)\right].$$

In a metal, we lose the high-lying spin-wave branch with $\omega_q\sim\Delta_c$.

We rewrite Eq. (2) as

$$\prod_{\mathrm{j}}(\omega-\Delta E_{\mathrm{j}})+A^2\chi_0{}^{+-}(\mathbf{q},0)\sum_{\mathrm{j}}|J_{\mathrm{j}}{}^+|^2\Delta N_{\mathrm{j}}\prod_{\mathrm{i}\neq\mathrm{j}}(\omega-\Delta E_{\mathrm{i}})=0,$$

where $\mathbf{j}$ specifies transitions between the sublevels of the f ion. At low temperatures, we have the level occupation numbers $N_{J-1}\ll 1$, $N_{J-2}\ll 1$, etc. Retaining only ΔN_j with $\mathbf{j}=(J,J-1)$, we find

$$(\omega-\Delta E_{J-1,J}-A^2\chi^{+-}_{\mathrm{j}}(q,0)\,\Delta N_{J,J-1}\cdot 2J)\prod_{\mathrm{j}\neq(J,J-1)}(\omega-\Delta E_{\mathrm{j}})=0. \tag{5}$$

In this temperature range, all the spin-wave branches except that corresponding to the transition between the lower levels are nondispersive, and their energy is equal to the difference between the energies $E_{m+1}-E_m$ of the f ion in the crystal field, the magnetic field, and the exchange field. The exchange field is produced by the conduction electrons. The expression in parentheses in (4) agrees at $T=0$ with Eq. (2) of Ref. 6. The derivation here proves our assertion in the Introduction regarding the range of applicability of this equation. At the same time, we see that the phenomenological anisotropy constant $B(T)$ introduced in Ref. 6, which determines the gap in the spectrum of the lower spin-wave branch, has the meaning of the difference between the low-lying energy levels of the f ion in the crystal field and the magnetic field:

$$B(T)=\Delta E_{J-1,J}-2AR_c{}^z.$$

The change in the occupation numbers N_J and N_{J-1} of the low-lying levels E_J and E_{J-1} at low temperatures is evidently also determined by spin waves:

$$\delta N_m=-\frac{T}{N}\sum_{\mathbf{q},n}\Bigg\{\frac{|J^{+}_{m+1,m}|^2\Delta N_{m+1,m}}{(i\omega_n-\Delta E_{m,m+1})(i\omega_n-\omega^{m,m+1}_{\mathbf{q}})}\times\prod_{\mathrm{i}\neq(m,m+1)}\frac{i\omega_n-\Delta E_{\mathrm{i}}}{i\omega_n-\omega_{\mathbf{q}}{}^{\mathrm{i}}}$$

$$+\frac{|J^{+}_{m,m-1}|^2\Delta N_{m,m-1}}{(i\omega_n-\Delta E_{m-1,m})(i\omega_n-\omega^{m-1,m}_{\mathbf{q}})}\times\prod_{\mathrm{i}\neq(m,m-1)}\frac{i\omega_n-\Delta E_{\mathrm{i}}}{i\omega_n-\omega_q{}^{\mathrm{i}}}\Bigg\}A^2\chi_0{}^{+-}(\mathbf{q},0).$$

Sine we have $N^0_J\approx 1$ and $N^0_{m\neq J}\approx 0$ at low temperatures (the superscript 0 means the "mean field"), we have

$$\delta N_J=-\frac{1}{N}\sum_{\mathbf{q}}\left[n_B(\omega_{\mathbf{q}}^{J-1,J})-n_B(\Delta E_{J-1,J})\right]$$

$$\approx-\frac{1}{N}\sum_{\mathbf{q}}n_B(\omega_{\mathbf{q}}^{J-1,J})$$

and thus

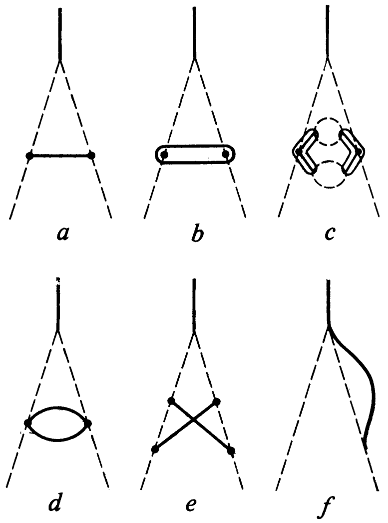
$$N_J=1-\frac{1}{N}\sum_{\mathbf{q}}n_B(\omega_{\mathbf{q}}),$$

$$N_{J-1}=\frac{1}{N}\sum_{\mathbf{q}}n_B(\omega_{\mathbf{q}}), \quad n_B(\omega)=[e^{\beta\omega}-1]^{-1},$$

and the average magnetization becomes

$$\langle J^z\rangle=JN_J+(J-1)N_{J-1}=J-\frac{1}{N}\sum_q n_B(\omega_q).$$

Equations (2) and (3) are incorrect if there are large corrections to the electron-spin vertex A. These corrections are of the form



In deriving an equation for the spin-wave spectrum,[6] Vedyaev and Nikolaev used for the vertex an equation based on a correction of type a as shown here (the solid line is a magnon line[6]). Diagram a is forbidden by spin conservation and does not occur (as was mentioned in Ref. 18). Diagram[1] b,

$$\sim\frac{A^2}{N}\sum_{\mathbf{k}}G_{\mathbf{k}}(i\omega)G_{\mathbf{k}+\mathbf{q}}(i\omega)\sum_{m,m'=-J}^{J}mm'(N_m\delta_{mm'}-N_mN_{m'}),$$

is exponentially small at low temperatures and does not contain a sum over the frequency. It is therefore also small in the "mean-field" temperature range. When we take (2) into account, we find that correction c becomes correction d, where the solid line without an oval represents the series analogous to (2) for the function

$$K_{\mathbf{ff}'}^{+-}(i\nu)=\frac{1}{N}\sum_{\mathbf{q}}e^{i\mathbf{q}(\mathbf{f}-\mathbf{f}')}\frac{K_0^{+-}(i\nu)}{1+A^2\chi_0^{+-}(\mathbf{q},i\nu)K_0^{+-}(i\nu)}.$$

In none of the three diagrams d, e, f can we single out a "resonant" electron-hole pair. These diagrams contain an integration over a broad range of the momentum, so that the estimate given in Ref. 18 remains valid for them; they can be discarded. Corrections b–f thus do not hurt Eq. (3).

3. EQUATIONS OF THE PHASE DIAGRAM

The bare Green's functions (4) correspond to the mean-field approximation, so that Eq. (3) is essentially a generalization of the Tyablikov equation to the case of an anisotropic s–f magnetic material. We can thus use (3) to determine such crude properties as the phase diagram. At $\omega_q \leqslant 0$ the ferromagnetic phase is unstable. Because of the flat regions on the Fermi surface, the electron susceptibility $\chi_0^{+-}(\mathbf{q},0)$ has a maximum at $\mathbf{q}\sim\mathbf{Q}$, so that Eq. (3) at $\omega=0$,

$$1+A^2\chi_0^{+-}(\mathbf{q},0)K_0^{+-}(0)=0, \tag{6}$$

gives us the phase diagram of a transition from a ferromagnetic state to a state with a helicoidal structure with a period $2\pi/Q$. The transition from the paramagnetic phase to the ferromagnetic phase is made less abrupt by a field, although at $h/T_c \ll 1$ a trace of the singularity remains. The effect of a field on an antiferromagnetic dielectric reduces to shifting T_N by $\sim h^2/T_c$, according to Ref. 19. This shift results from a Zeeman shift of the levels of the magnetic ions. In metals, a field also pushes the Fermi surfaces of electrons with different spin projections further apart. Following Ref. 12, we classify all electrons as either "resonant" electrons, which occupy the flat parts of the Fermi surface, or "nonresonant" electrons, which occupy the remainder of the Fermi surface, which we will assume to be spherical. The nonresonant electrons are insensitive to the field since $h/\mu \ll 1$. We emphasize that the resonance condition and, correspondingly, the condition for a transition from a paramagnet to a helicoidal structure may change. These changes can be followed by analyzing the poles of the longitudinal Green's function in the paramagnetic phase:

$$T_c+A^2\chi^{zz}(\mathbf{q}=0)K^{zz}=0, \tag{7}$$

$$T_N+A^2\chi^{zz}(\mathbf{q}=\mathbf{Q})K^{zz}=0, \tag{8}$$

$$\chi^{zz}(\mathbf{q})=\frac{1}{N}\sum_{\mathbf{k}\sigma}\frac{n(\xi_{\mathbf{k}+\mathbf{q}}^{\sigma})-n(\xi_{\mathbf{k}}^{\sigma})}{\xi^{\sigma}_{\mathbf{k}+\mathbf{q}}-\xi_{\mathbf{k}}^{\sigma}},$$

$$K^{zz}=\sum_{mm'}(\delta_{mm'}m^2-mm'N_{m'})N_m.$$

Equation (7) gives the condition for a transition (albeit smoothed over the field) to the ferromagnetic phase, while Eq. (8) gives the condition for a transition to a helicoidal structure.

If there is no anisotropy and no external field, we find from (7) the usual expression for $T_c=2A^2g_0J(J+1)/3$, while in the Ising limit, $D_{2n}^0\to\infty$, we have $T_c=2A^2g_0J^2$; i.e., the anisotropy cannot change T_c by a factor greater than three. Ignoring quantities $\sim T/\mu$ and using the conditions $\Delta/2\mu \ll 1$ and $q/2p_0<1$, we find, with an accuracy to $(\Delta/2\mu)^3$,

$$\chi_0^{+-}(\mathbf{q},0)=\chi_0^{zz}(\mathbf{q})=g_0[1-(q/2p_0)^2], \qquad g_0=v_cp_0m/2\pi^2,$$

where m is the electron mass, v_c is the volume of the unit cell, p_0 is the Fermi momentum, and g_0 is the state density on the nonresonant Fermi surface. To calculate the resonant contribution, we replace the Fermi function by a step with an angle $\tan\tilde{\alpha}\sim 1/2T$. We use the mean value theorem to integrate over the flat region. As a result, we find that the susceptibility χ^{+-} is proportional to the quantity

$$\chi_0^{+-}(\mathbf{q},0)\propto\ln\left|\frac{(q+Q^+)(q-Q^-)-2\alpha q}{(q-Q^+)(q-Q^-)+2\alpha q}\right|,$$

$$Q^{\pm}=p_++p_-, \qquad p_{\pm}=[2m\mu^*(1\pm\Delta_c/2\mu^*)]^{1/2}, \qquad \alpha=(2mT)^{1/2},$$

where μ^* is the chemical potential, which is reckoned from the critical point on the Fermi surface.[20]

In the limit $T\to 0$, the quantity $\chi_0^{+-}(q,0)$ diverges logarithmically at the point p_++p_-, as was mentioned in Ref. 12 (see also the Conclusion). At a nonzero temperature the susceptibility has a maximum

$$\chi_{\text{res}}^{+-}(Q,0)=-\frac{v_c}{(2\pi)^3}\frac{m\Delta S_{\text{res}}}{Q}\ln\frac{\alpha}{Q} \tag{9}$$

at $Q=p_++p_-$ (a calculation of the susceptibility in the case of the frequently used spectral property $\varepsilon_{\mathbf{k}}=-\varepsilon_{\mathbf{k}+\mathbf{Q}}$, leads to the same logarithmic dependence on the temperature[21]). After corresponding calculations for the longitudinal susceptibility we find that this susceptibility has a maximum

$$\chi_{\text{res}}^{zz}(Q)=-\frac{v_c}{(2\pi)^3}\frac{m\Delta S_{\text{res}}}{Q}\ln\frac{\alpha}{2p_-} \tag{10}$$

at the point $Q=2p_-$, where ΔS_{res} is the area of the resonant part of the Fermi surface. By solving Eq. (6) jointly with the equation for the magnetization in the external field we can thus find the temperature dependence of the vector of the helicoidal structure. The effective chemical potential μ^* can be found if a single experimental point is known.

The system can change from the paramagnetic phase to a helicoidally ordered phase if

$$\chi^{zz}(\mathbf{Q})\geqslant\chi^{zz}(0).$$

Analysis of this condition reveals that it holds only for a certain area of the resonant surface:

$$\Delta S_{\text{res}}>3(Q/2p_0)^3S,$$

where S is the area of the nonresonant part of the Fermi surface.

Using the electron susceptibilities calculated in this manner, we can very easily take into account the important features of the Fermi surface and also the temperature and field dependence of $\chi(\mathbf{q})$.

Nevertheless, it is not possible at present to calculate phase diagrams for specific rare earth metals, since the data available (on the single-ion anisotropy constants, for example) not only differ greatly in magnitude but also differ in sign.[3,4,9] We will now show that certain microscopic constants can be determined from measurements in a low-symmetry phase.

4. TRANSITION FROM A FERROMAGNETIC HELIX TO A FERROMAGNET

Let us consider the opposite transition—from a helicoidal structure to a ferromagnet—in an external magnetic field. We restrict the discussion to the case of the ferromagnetic helix which is observed in holmium and erbium. We will derive an equation for the transition point, and we will find the low-temperature behavior of the magnetization components near this point. For this purpose, we write a system of equations for the Green's functions in the self-consistent field approximation:

$$G^{\alpha\beta}_{\mathbf{kk}'}=\langle T\overset{+}{\psi}_{\mathbf{k}\alpha}(\tau)\psi_{\mathbf{k}'\beta}(\tau')\rangle,$$

where we have $\psi_{\mathbf{k}\alpha}=(c_{\mathbf{k}\alpha}, c_{\mathbf{k}+\mathbf{Q}\alpha})$ in the ferromagnetic-helix phase, and **Q** is the vector of the magnetic structure. The crystal is inhomogeneous, and the solution of the system of equations is the following matrix Green's function:

$$G^{\alpha\beta}_{\mathbf{kk}'}=\frac{\mathscr{A}_{\alpha\beta}(v_1^2, R_\perp^f)}{i\omega-\omega_1^+}+\frac{\mathscr{A}_{\alpha\beta}(u_1^2, -R_\perp^f)}{i\omega-\omega_1^-}+\frac{\mathscr{B}_{\alpha\beta}(v_2^2, R_\perp^f)}{i\omega-\omega_2^+}+\frac{\mathscr{B}_{\alpha\beta}(u_2^2, -R_\perp^f)}{i\omega-\omega_2^-}, \tag{11}$$

where the matrices $\hat{\mathscr{A}}$ and $\hat{\mathscr{B}}$ are

$$\hat{\mathscr{A}}(v^2, R_\perp^f)=\begin{pmatrix} v^2 & 0 & 0 & AR_\perp^f/2\nu_1 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ AR_\perp^f/2\nu_1 & 0 & 0 & v^2 \end{pmatrix},$$

$$\hat{\mathscr{B}}(v^2, R_\perp^f)=\begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & v^2 & AR_\perp^f/2\nu_2 & 0 \\ 0 & AR_\perp^f/2\nu_2 & v^2 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix};$$

$$v_1^2=\frac{1}{2}\left(1-\frac{\xi_{\mathbf{k}+\mathbf{Q}}-\xi_{\mathbf{k}}+\Delta}{\nu_1}\right),$$

$$v_2^2=\frac{1}{2}\left(1-\frac{\xi_{\mathbf{k}}-\xi_{\mathbf{k}+\mathbf{Q}}+\Delta}{\nu_2}\right),\quad u_i^2+v_i^2=1,$$

$$\omega_i^\pm=\frac{1}{2}(\xi_{\mathbf{k}+\mathbf{Q}}+\xi_{\mathbf{k}}\pm\nu_i),\qquad \nu_1=[(\xi_{\mathbf{k}+\mathbf{Q}}-\xi_{\mathbf{k}}+\Delta)^2+(AR_\perp^f)^2]^{1/2},$$

$$\nu_2=[(\xi_{\mathbf{k}+\mathbf{Q}}-\xi_{\mathbf{k}}-\Delta)^2+(AR_\perp^f)^2]^{1/2},\quad \Delta=2AR_\parallel^f+g_ch,$$

and $R_\parallel^f$ and $R_\perp^f$ are the average values of the magnetization along the z axis and in the plane. In order to find $R_\parallel^f$ and $R_\perp^f$ we would generally have to diagonalize a matrix of rank $(2J+1)$, but this is not necessary in the present case since the transverse component of the magnetization is small near the transition point. Making use of this circumstance, we find the wave functions and energies of the levels of the f ion by perturbation theory. The expressions for $R_\parallel^f$ and $R_\perp^f$ in the phase of the ferromagnetic helix are then found to be $R_\parallel^f\propto(h_c-h)$, while the transverse components exhibits a square-root behavior ($R_\perp^f\propto(h_c-h)^{1/2}$). This temperature dependence of the structure vector **Q** describes the correct tendency toward an increase with increasing temperature.

The phase diagram which can be constructed from our equations is valid only if the fluctuational region is narrow. As we move

$$R_\parallel^f=\sum_m\{[m+(AR_\perp^c)^2((m-1)\gamma_{m-1}+(m+1)\gamma_{m+1})]\times\exp(-\beta E_m^*)\}/Z_0, \tag{12}$$

$$R_\perp^f=2AR_\perp^c\sum_m(\beta_{m-1}+\beta_{m+1})\exp(-\beta E_m^*)/Z_0,$$

$$Z_0=\sum_m[1+(AR_\perp^c)^2(\gamma_{m-1}+\gamma_{m+1})]\exp(-\beta E_m^*), \tag{13}$$

$$E_m^*=E_m+(AR_\perp^c)^2(\beta_{m-1}+\beta_{m+1}),$$

$$\gamma_{m+1}=|\overset{+}{J}_{m+1,m}|^2/\Delta E^2_{m+1,m},\qquad \beta_{m+1}=\Delta E_{m+1,m}\gamma_{m+1}.$$

We find expressions for the $R_\parallel^c$ and $R_\perp^c$ components of the magnetization of the c subsystem from the Green's functions which we have found, (12):

$$R_\parallel^c=\frac{1}{N}\sum_{\mathbf{k}}\{v_1^2n(\omega_1^+)+u_1^2(\omega_1^-)-v_2^2n(\omega_2^+)-u_2^2n(\omega_2^-)\}, \tag{14}$$

$$R_\perp^c=\frac{AR_\perp^f}{N}\sum_{\mathbf{k}}\frac{n(\omega_1^+)-n(\omega_1^-)}{\nu_1}. \tag{15}$$

The system goes into the ferromagnetic state in the limit $R_\perp^f\to0$. Substituting (15) into (13), and letting $R_\perp^f$ tend to zero, we find an equation which is the same as that derived earlier for the transition from the ferromagnetic phase, Eq. (6).

Let us find the critical transition field h_c at low temperatures. Near h_c, the magnetization of the c subsystem can be written

$$R_\parallel^c=A^2\chi_0^{+-}(0, 0)R_\parallel^f.$$

Using this result, we find the following expression for h_c:

$$h_c=2A^2J[\chi_0^{+-}(\mathbf{Q}, 0)-\chi_0^{+-}(0, 0)]-\Delta D,$$

$$\Delta D=\sum_{n=1}^{3}D_{2n}^0[J^{2n}-(J-1)^{2n}].$$

The longitudinal magnetization component $R_\parallel^f$ behaves linearly near h_c in the ferromagnetic helix, while the behavior of the transverse component is a square-root behavior:

$$R_\parallel^f=J-\frac{h_c-h}{2A^2\chi_0^{+-}(\mathbf{Q},0)}, \tag{16}$$

$$R_\perp^f=[2\Delta E_{J-1,J}/(A^2\chi_0^{+-}(\mathbf{Q}, 0))^2]^{1/2}(h_c-h)^{1/2}. \tag{17}$$

By studying the field dependence of the magnetization experimentally we can determine the quantity $A^2\chi_0^{+-}(\mathbf{Q}, 0)$ and also the energy of the transition between the low-lying levels of the f ion. If we find the gap in the spin-wave spectrum in a magnetic field $h > h_c$ at low temperatures, we can then use Eqs. (16) and (17) to find the distance the low-lying levels are moved apart by the single-ion anisotropy ΔD.

5. CONCLUSION

This theory can be used to describe a rare earth metal which has either an easy-axis anisotropy (Ho below 20 K or Er) or an easy-plane anisotropy (Tb, Dy, Ho above 20 K, and Tm) in a strong longitudinal ($h \parallel c$) magnetic field. It follows from an analysis of Eq. (3) for the spectrum of spin waves and the poles of the longitudinal susceptibility that the critical field for the transition from the ferromagnet to the helicoidal structure depends strongly on the area of the flat part of the Fermi surface. Detailed measurements of the field dependence of the magnetization of a rare earth metal near the transition might therefore yield information on this area. According to our equations, as the field is varied away from the ferromagnetic helix the longitudinal magnetization component exhibits a linear behavior ($R^f_\parallel \propto (h_c - h)$), while the transverse components exhibits a square-root behavior ($R^f_\perp \propto (h_c - h)^{1/2}$). This temperature dependence of the structure vector **Q** describes the correct tendency toward an increase with increasing temperature.

The phase diagram which can be constructed from our equations is valid only if the fluctuational region is narrow. As we move away from the ferromagnetic region the dimension of the fluctuations is cut off by the field. The role played by fluctuations in a helicoidal structure in a rare earth metal was studied in Refs. 22 and 23, where it was shown that the transitions may be of either first or second order. So far, there has been no study of the case with a pronounced anisotropy and an external magnetic field. Other open questions are the mechanisms which lead to the sequence of phase transitions in easy-plane rare earth metals and the roles played by the Coulomb and phonon interactions. These interactions will obviously not be unaffected by ribbon singularities on the Fermi surface. It is surprising that no space-charge waves or transitions of the displacement type have been found in rare earth metals, since the constant of the Coulomb interaction (either the *s-s* or *s-f* interaction) is usually larger than the constant of the *s-f*-exchange interaction. The existence of, say, space-charge waves might cast light on the nature of the *z* helix in Er, where the angular momenta of the *f* ions are directed along the *c* axis and vary in magnitude along this axis in the temperature range 53.5–85 K. If there is a space-charge wave, then the crystal field sensed by each *f* ion will differ from that sensed by the adjacent *f* ion along the *c* axis. The angular momentum of the *f* ion will then be determined primarily by the state which is the lower state in the given crystal field, and this state will not necessarily have the maximum possible projection. It thus seems to us to be extremely important to seek space-charge waves and displacement transitions in rare earth metals.

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APPENDIX

We will show here that the procedure of Callen and Callen is not appropriate for a rare earth metal. We write the free energy of the system as the sum of the energies of isotropic and anisotropic parts. We write the anisotropic component as

$$\varepsilon_{\text{an}} = \sum_{l,m} C_l{}^m Y_l{}^m(\vartheta, \varphi), \tag{A.1}$$

where the angles ϑ and φ specify the direction of the saturation magnetization with respect to some selected frame, usually the crystallographic axes. The coefficients C_l^m depend on external parameters such as the pressure, the temperature, and the magnetic field; in general, they also depend on the magnitude of the magnetization vector. To calculate the C_l^m we take the following approach: Since a rare earth metal has a strong spin-orbit coupling ($\sim 10^3$–10^4 cm^{-1}; Refs. 1, 3, and 4), the various J multiplets of the *f* ions are separated by a large energy gap, and we need consider only the lowest-lying multiplet. We can thus use the Wigner-Eckart theorem and write the Hamiltonian of the single-ion anisotropy in terms of equivalent Stephens operators $\hat{O}$ constructed from the components of the total angular momentum operators J_f at the site *f*:

$$H_{\text{an}} = \sum_{f} D_n{}^m O_n{}^m(\mathbf{J}_f). \tag{A.2}$$

Here the D_n^m are the microscopic anisotropy constants.[4] The operators $\hat{O}$ are written in the intrinsic representation of the operator J^z, where the *z* axis runs parallel to the hexagonal axis of the rare-earth crystal. In the Callen and Callen procedure, after transforming in (A.2) to a quantization axis along the magnetization **M** of the crystal and after thermodynamic averaging, a cylindrical symmetry about **M** is assumed explicitly. The free energy is then written in the form

$$F = F_0 + \sum_{l,m} K_l{}^m(0) \langle Y_l{}^0(\mathbf{J}) \rangle Y_l{}^m(\vartheta, \varphi), \tag{A.3}$$

where the Y_l^m are the spherical harmonics. Comparison of (A.3) with (A.1) gives us the dependence of C_l^m on the parameters of the system. The averaging of the operators $Y_1(J)$ is usually carried out with a distribution function $\propto \exp(-xm)/\Sigma_{m'} \exp(-xm')$. This procedure corresponds to an approximation of the mean-field type with an equidistant *f*-ion spectrum. Under what conditions are the magnetic-anisotropy effects described completely by expressions (A.1) and (A.3)? The Hamiltonian of the system is

$$H = H_{\text{an}} + H_0 + H_1, \qquad H_0 = -2A(0) \sum_{f} \langle \mathbf{J} \rangle \mathbf{J}_f, \tag{A.4}$$

where H_0 describes the change interaction in the mean-field approximation, and H_1 describes some spherically symmetric exchange interaction (an *s-f*-exchange interaction or a Heisenberg interaction) minus the mean field. From (A.4) we have the following expression for the free energy;

$$\beta F=\int_0^\beta d\lambda\langle H\rangle_\lambda=\beta(F_{\text{an}}+F_0+F_1),$$

$$\beta F_i=\int_0^\beta d\lambda\,\mathrm{Sp}\{H_i\exp(-\lambda H)\}/\mathrm{Sp}\{\exp(-\lambda H)\},\quad i=\text{an},\ 0,\ 1. \tag{A.5}$$

We see from (A.5) that if F_{an} is to completely describe the anisotropy effects then we must discard the Hamiltonian H_{an} in the Gibbs distribution function in F_0 and F_1. On the other hand, if in the expansion

$$\langle Y_l^m(\mathbf{J})\rangle=\sum_{m'}b_l(m,m')\langle Y_l^{m'}(\mathbf{J})\rangle$$

we wish to retain only the terms with $m'=0$, i.e., $b_l(m,0)=Y_l^m(\vartheta,\varphi)$, then we must also discard this Hamiltonian in F_{an}, so that we are actually left with a Hamiltonian which is spherically symmetric. The primary requirement is thus the requirement that H_{an} cause only a slight perturbation of the distribution of levels determined by H_0+H_1. The magnetic-ordering temperatures in heavy rare earths are[1,3,4]

$$T_c\sim 2A^2g_0J(J+1)/3\sim 10^2\ \text{cm}^{-1}$$

corresponding to a distance $\sim$10–50 cm^{-1} between single-ion magnetic sublevels (for Er, for example, we have $J=15/2$). The magnetic anisotropy energy is[1,3,4] ~ 10–10^2 cm^{-1}, so that we cannot construct a perturbation theory in the energy parameters; correspondingly, even phenomenological expression (A.1) cannot be used.

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1) When the Holstein–Primakoff representation $J_f^zc^+c\to(J-a_f^+a_f)c^+c$ is used,[18] the corrections of type b and other corrections containing ovals with several disconnected blocks (corrections to the occupation numbers) do not appear at all.

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