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Spin-density-wave state of chromium

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A Landau theory of the spin-density-wave (SDW) state in chromium alloys is constructed. In this theory the cubic symmetry of the bcc lattice and the symmetry with respect to rotations in spin space of the exchange interaction are manifest in the free-energy expansion. The Landau coefficients are first obtained for a general two-band model as sums over the Matsubara frequencies, then evaluated near the triple point for a simple class of Fermi surfaces. The competition of the various polarization and wave-vector states is studied, and the stable phase of each of the models determined. It is found that for a Fermi surface consisting of two spheres of unequal radii the triple-$\vec{Q}$ state is the stable one, whereas the single-$\vec{Q}$ state is stable for an octahedral Fermi surface. The direction of the wave vector of the SDW is determined, for the octahedral model, to be in one of the $\langle 100 \rangle$ directions, in agreement with the experimental result for pure chromium.

I. INTRODUCTION

A. The nature of the system

Chromium is a body-centered-cubic metal which undergoes a transition at 312 K to a state in which the electrons form a static spin-density wave (SDW) with a period which is incommensurate with the lattice. The magnetized state consists of domains, each containing a single wave with wave vector $\vec{Q}$ along one of the cube axes. In the range of temperature from the Néel temperature 312 K (T_N) to the spin-flip temperature 123 K (T_F), the wave is transversely polarized, with the magnetization pointing along one of the other cube axes. Below 123 K the wave is longitudinally polarized.

The discovery that chromium is antiferromagnetic was made by Shull and Wilkinson[1] in 1953 as a result of a neutron-diffraction experiment. Further neutron-diffraction studies[2-6] have yielded the above picture. The results of Werner, Arrott, and Kendrick[7] show a small jump in the magnetization at T_N, which is often interpreted as being due to a first-order transition.

Besides the fundamental SDW, there have been observed in chromium other structures associated with the wave. In particular, there is a charge-density wave (CDW) of wave vector $2\vec{Q}$, as shown by the x-ray scattering results of Tsunoda *et al*,[8] and also a SDW of wave vector $3\vec{Q}$, as shown by the neutron-scattering results of Pynn *et al.*[9] Also, there is some evidence, as found in a comparison of the x-ray-and neutron-scattering intensities made by Eagen and Werner,[10] for the existence of a lattice strain wave (SW) of wave vector $2\vec{Q}$.

B. Theories of the SDW state

The theory of the SDW state is founded on the work of Overhauser,[11] who showed that the paramagnetic state of an electron gas with a uniform background of positive charge, when treated in the Hartree-Fock approximation, was unstable with respect to the formation of a static spin-density wave. Overhauser pointed to the magnetism of chromium as an example of the SDW state, and explained several experimental observations in terms of a single band model. A more accurate description of the SDW state in chromium has since been the object of numerous studies.[12-22]

The actual mechanism operating in chromium to produce the SDW has turned out to be the coincidence of large portions of the Fermi surface from two different bands under translation of one band by the wave vector $\vec{Q}$ of the SDW, as suggested by Lomer.[23]

Fedders and Martin[13] studied a model with a

Fermi surface consisting of an electron sphere and a hole sphere of equal radius. A model Fermi surface consisting of spheres of unequal radii was studied by Rice[15] and by Malaspinas and Rice.[16] This model took account of the imperfect nesting of the actual Fermi surface of Cr, and allowed the study of the effects of alloying, where the radii of the spheres were assumed to change with varying alloy concentration. Shibatani *et al.*[17] introduced a model with perfect octahedra as the Fermi surface, more closely resembling that of chromium. Nakanishi and Maki[18] considered the transition from the paramagnetic to the incommensurate SDW state using the unequal sphere model band structure, making an expansion in powers of the departure of the wave vector away from the commensurate one, valid near the triple point. They neglected, however, the effect of the finite electron reservoir in actual chromium. This neglect changes their conclusions since Malaspinas and Rice[16] obtain a second-order transition in contrast to the first-order transition found by Nakanishi and Maki. Fenton[19,20] also has considered this transition. He has used the octahedron band structure and allowed the magnetization density to contain three Fourier waves, with wave vectors mutually perpendicular. The free energy was lowest when just a single wave had a nonzero amplitude, thus proving the stability of a single-$\vec{Q}$ state within the model.

This paper unifies and completes previous mean-field theories of the SDW state. In particular, there is a full treatment of the relation of the shape of the Fermi surface to the nature of the condensed phase. The theory is an improvement over previous theories firstly in that all in a general class of Fermi surfaces (those consisting of an electron part and a hole part of the same shape) are considered simultaneously. Secondly, the condensate is allowed complete freedom to contain several Fourier components of magnetization, each of arbitrary polarization. In Sec. II, a series of steps leads from a two-band Hamiltonian for the electron assembly to expressions for the general second- and fourth-order Landau coefficients in an expansion of the free energy in powers of the magnetization density. In Sec. III, the competition of magnetized states, depending on the relative magnitudes of the Landau coefficients, is discussed. In Sec. IV, specific band structures are considered, giving predictions on the stability of various states for the models. The paper ends in Sec. V with a discussion of the results.

II. MICROSCOPIC DERIVATION OF THE FREE-ENERGY COEFFICIENTS

A. Introduction

The variational principle of statistical mechanics, as discussed, e.g., by Huber,[24] provides a means for obtaining a Landau expansion for a system described by a Hamiltonian. The form of the theorem most useful is that the grand potential $\Omega(T,\mu)$, where μ is the chemical potential, is always less than a certain class of trial potentials containing variational parameters. The coefficients of the expansion are known in terms of the microscopic Hamiltonian, and the variational parameters are identified with expectation values of quantum-mechanical operators.

The aim of this section is to obtain expressions for the coefficients in a Landau expansion describing the transition from the paramagnetic to the incommensurate SDW state. The variational method is applied to the system of two bands of electron states (a,b) in exchange interaction, and a third band (c) acting as a finite reservoir.

B. Variation of the grand potential

The minimum principle for the grand potential $\Omega(T,\mu)$ of a system described by a Hamiltonian H is that $\Omega(T,\mu)$ is less than or equal to any trial potential $\Omega_t(T,\mu)=\langle H-\mu N\rangle_t-TS_t$, where $\langle\ \rangle_t$ denotes the average calculated with respect to a trial density operator ρ_t.

Following Rice,[15] the model Hamiltonian describing the system is taken to be

$$H=\sum_{\substack{k\sigma\\ \alpha=abc}}\epsilon_{\alpha k}c^{\dagger}_{\alpha k\sigma}c_{\alpha k\sigma}+\bar{v}\sum_{\substack{kk'q\\ \sigma\sigma'}}a^{\dagger}_{k+q\sigma}b^{\dagger}_{k'-q\sigma'}b_{k'\sigma'}a_{k\sigma}$$

$$=H_0+H'\ .$$

Bands a and b are the nesting bands composed of Bloch states for electrons in the lattice potential, interacting to form the SDW. Band c represents the other electrons, acting merely as a finite electron reservoir. $\bar{v}$ is the average of the effective exchange interaction over energies of importance for the interaction of the bands. For the present the form of the bands $\epsilon_{\alpha k}$ is left unspecified.

The trial Hamiltonian is assumed to have the form

$$H_t=H_0+\sum_{\substack{kq_r\\ \sigma\sigma'}}[\Delta_{\sigma\sigma'}(q)a^{\dagger}_{k+q\sigma}b_{k\sigma'}+H.a.]\ , \qquad (1)$$

where the variational parameters $\Delta_{\sigma\sigma'}(q)$ are arbitrary complex numbers, and where $\sum_{q_r}$ denotes a summation over q restricted to a set of wave vectors q_r. In order that the trial Hamiltonian retain the cubic symmetry of the high-temperature phase of chromium, the $\vec{q}_r$ will later be taken to be $\pm\vec{q}_i$ ($i=1,2,3$), where the $\vec{q}_i$ are three mutually orthogonal vectors.

Variation of the $\Delta_{\sigma\sigma'}(q)$ leads to a variation in Ω_t of

$$\delta\Omega_t=-\sum_{q_r\sigma\sigma'}[\Delta_{\sigma\sigma'}(q)+\bar{v}\psi_{\sigma\sigma'}(q)]\delta\psi^*_{\sigma\sigma'}(q) + \text{c.c.}, \qquad (2)$$

where

$$\psi_{\sigma\sigma'}(q)=\sum_{k'}\langle b^\dagger_{k'-q\sigma'}a_{k'\sigma}\rangle\ .$$

Restriction to the SDW state requires setting

$$\Delta_{\sigma\sigma'}(q)=(\vec{M}_q\cdot\vec{\sigma})_{\sigma\sigma'}$$

with $\vec{M}_{-q}=\vec{M}^*_q$.

Equation (2) gives an expression for the grand potential in terms of microscopic quantities. It is used in the appendix to obtain microscopic expressions for the phenomenological free-energy coefficients of Sec. II D. Before proceeding to the phenomenological discussion, however, a brief aside is made on the accuracy of the expansion.

C. Truncation of the expansion of $\delta\Omega_t$

When calculating $\delta\Omega_t$ with the trial Hamiltonian given by Eq. (1), besides the expectation values $\psi_{\sigma\sigma'}(q)$ there are others arising from higher-order perturbation terms, so that $\delta\Omega_t$ should be written

$$\delta\Omega_t=-\sum_{q_r\sigma\sigma'}[\Delta_{\sigma\sigma'}(q)+\bar{v}\psi_{\sigma\sigma'}(q)]\delta\psi^*_{\sigma\sigma'}(q) + \text{c.c.}+\sum_e \bar{v}\psi_e\delta\psi^*_e\ ,$$

where the ψ_e are extra nonzero expectation values, e.g., $\psi_e=\psi^a_{\sigma\sigma}(2q)=\sum_k\langle a^\dagger_{k+2q\sigma}a_{k\sigma}\rangle$. To take these contributions properly into account requires that further terms be added to the trial Hamiltonian, giving extra terms $\Delta_e\delta\psi^*_e$ in $\delta\Omega_t$. For an expansion consistent to fourth order in $\Delta_{\sigma\sigma'}(q)$, the only such terms which are necessary are the $2q$ CDW harmonics. However, an accurate theory taking charge-density harmonics into account requires that the effects of phonons be included, and as such are not included, the terms involving CDW components are also omitted.

D. Phenomenological grand potential expansion

So far the number of components of the order parameter has been left unspecified as $\{\vec{M}_{q_r}\}$. Group theory may be used to determine the minimum number of components required in order that the theory be compatible with the symmetry of the system.[25,26] Assume for definiteness that the magnetization of Cr consists of a single Fourier component of magnetization $\vec{M}_{q_1}$, with $\vec{q}_1$ directed along (1,0,0), for example. In order to conform to the bcc symmetry of paramagnetic chromium, components of magnetization with wave vectors obtained by symmetry operations of the lattice acting on $\vec{q}_1$ (forming the star of $\vec{q}_1$) must be included in the description on an equal footing with those of $\vec{q}_1$. The symmetry group of the paramagnetic lattice is Im3m. The star of $\vec{q}_1$ consists of the wave vectors $\pm\vec{q}_1$, $\pm\vec{q}_2$, and $\pm\vec{q}_3$, along (1,0,0), (0,1,0), and (0,0,1), respectively. Therefore the order parameter consists of 18 real numbers formed from $\vec{M}_i,\vec{M}_{\bar{i}}$, with $i=1,2,3$. (Here $\bar{i}$ denotes $\vec{q}_{\bar{i}}=-\vec{q}_i$.)

The grand potential is expanded as the sum of terms formed from the $\vec{M}_i$, each term invariant under the symmetry operations of the system. As there is no spin-orbit coupling in the model, the system is invariant with respect to separate infinitesimal rotations in spin space and rotations of the cubic group in $\vec{k}$ space. Spin-space rotations force the second-order invariants to consist of sums of terms of the form $\vec{M}_p\cdot\vec{M}_q$. Invariance of the Hamiltonian for the electron system under translation of a lattice vector, together with the assumption that the periodicity of the magnetism is incommensurate with that of the lattice, requires that $p=\bar{q}$. The fourth-order invariants must similarly consist of sums of terms of the form $(\vec{M}_p\cdot\vec{M}_q)(\vec{M}_r\cdot\vec{M}_s)$ with $\vec{k}_p+\vec{k}_q+\vec{k}_r+\vec{k}_s=0$. Invariance under time reversal eliminates terms containing an odd number of $\vec{M}_q$. A set of generators for the cubic group O_h is $C_4(001)$, $C_3(111)$, and i (inversion). Under these operations, the order parameter transforms as

$$\begin{array}{llll}
C_4(001)\colon & M_{1x}\to M_{2y}, & C_3(111)\colon\ M_{1x}\to M_{2y}, & i\colon\ M_{1x}\to M_{\bar{1}x},\\
& M_{1y}\to -M_{2x}, & M_{1y}\to M_{2z}, & M_{1y}\to M_{\bar{1}y},\\
& M_{1z}\to M_{2z}, & M_{1z}\to M_{2x}, & M_{1z}\to M_{\bar{1}z},\\
& M_{2x}\to M_{\bar{1}y}, & M_{2x}\to M_{3y}, & M_{2x}\to M_{\bar{2}x},\\
& M_{2y}\to -M_{\bar{1}x}, & M_{2y}\to M_{3z}, & M_{2y}\to M_{\bar{2}y},\\
& M_{2z}\to M_{\bar{1}z}, & M_{2z}\to M_{3x}, & M_{2z}\to M_{\bar{2}z},\\
& M_{3x}\to M_{3y}, & M_{3x}\to M_{1y}, & M_{3x}\to M_{\bar{3}x},\\
& M_{3y}\to -M_{3x}, & M_{3y}\to M_{1z}, & M_{3y}\to M_{\bar{3}y},\\
& M_{3z}\to M_{3z}, & M_{3z}\to M_{1x}, & M_{3z}\to M_{\bar{3}z}.
\end{array}$$

The only second-order expression, invariant with respect to rotations in spin space, translations in real space, and rotations of the cubic group in real space is

$$\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_{\bar{1}}+\vec{\mathbf{M}}_2\cdot\vec{\mathbf{M}}_{\bar{2}}+\vec{\mathbf{M}}_3\cdot\vec{\mathbf{M}}_{\bar{3}}\ .$$

There are four fourth-order invariants. They are

$$\begin{aligned}
&(\mathrm{a})\ \ (\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_{\bar{1}})^2+(\vec{\mathbf{M}}_2\cdot\vec{\mathbf{M}}_{\bar{2}})^2+(\vec{\mathbf{M}}_3\cdot\vec{\mathbf{M}}_{\bar{3}})^2\ ,\\
&(\mathrm{b})\ \ (\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_1)(\vec{\mathbf{M}}_{\bar{1}}\cdot\vec{\mathbf{M}}_{\bar{1}})+(\vec{\mathbf{M}}_2\cdot\vec{\mathbf{M}}_2)(\vec{\mathbf{M}}_{\bar{2}}\cdot\vec{\mathbf{M}}_{\bar{2}})+(\vec{\mathbf{M}}_3\cdot\vec{\mathbf{M}}_3)(\vec{\mathbf{M}}_{\bar{3}}\cdot\vec{\mathbf{M}}_{\bar{3}})\ ,\\
&(\mathrm{c})\ \ (\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_{\bar{1}})(\vec{\mathbf{M}}_2\cdot\vec{\mathbf{M}}_{\bar{2}})+(\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_{\bar{1}})(\vec{\mathbf{M}}_3\cdot\vec{\mathbf{M}}_{\bar{3}})+(\vec{\mathbf{M}}_2\cdot\vec{\mathbf{M}}_{\bar{2}})(\vec{\mathbf{M}}_3\cdot\vec{\mathbf{M}}_{\bar{3}})\ ,\\
&(\mathrm{d})\ \ (\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_2)(\vec{\mathbf{M}}_{\bar{1}}\cdot\vec{\mathbf{M}}_{\bar{2}})+(\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_3)(\vec{\mathbf{M}}_{\bar{1}}\cdot\vec{\mathbf{M}}_{\bar{3}})+(\vec{\mathbf{M}}_2\cdot\vec{\mathbf{M}}_3)(\vec{\mathbf{M}}_{\bar{2}}\cdot\vec{\mathbf{M}}_{\bar{3}})+(\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_{\bar{2}})(\vec{\mathbf{M}}_{\bar{1}}\cdot\vec{\mathbf{M}}_2)\\
&\qquad\quad +(\vec{\mathbf{M}}_1\cdot\vec{\mathbf{M}}_{\bar{3}})(\vec{\mathbf{M}}_{\bar{1}}\cdot\vec{\mathbf{M}}_3)+(\vec{\mathbf{M}}_2\cdot\vec{\mathbf{M}}_{\bar{3}})(\vec{\mathbf{M}}_{\bar{2}}\cdot\vec{\mathbf{M}}_3)\ .
\end{aligned}$$

Time-reversal invariance eliminates terms with an odd number of $\vec{\mathbf{M}}_q$.

The form which the expansion of the grand potential must take can now be written down:

$$\begin{aligned}
\Omega_t=\Omega_0&+A\sum_i(\vec{\mathbf{M}}_i\cdot\vec{\mathbf{M}}_{\bar{\imath}})+u_1\sum_i(\vec{\mathbf{M}}_i\cdot\vec{\mathbf{M}}_{\bar{\imath}})^2+u_2\sum_i(\vec{\mathbf{M}}_i\cdot\vec{\mathbf{M}}_i)(\vec{\mathbf{M}}_{\bar{\imath}}\cdot\vec{\mathbf{M}}_{\bar{\imath}})+u_3\sum_{i>j}(\vec{\mathbf{M}}_i\cdot\vec{\mathbf{M}}_{\bar{\imath}})(\vec{\mathbf{M}}_j\cdot\vec{\mathbf{M}}_{\bar{\jmath}})\\
&+u_4\sum_{i>j}[(\vec{\mathbf{M}}_i\cdot\vec{\mathbf{M}}_j)(\vec{\mathbf{M}}_i\cdot\vec{\mathbf{M}}_{\bar{\jmath}})+(\vec{\mathbf{M}}_i\cdot\vec{\mathbf{M}}_{\bar{\jmath}})+(\vec{\mathbf{M}}_i\cdot\vec{\mathbf{M}}_{\bar{\jmath}})(\vec{\mathbf{M}}_{\bar{\imath}}\cdot\vec{\mathbf{M}}_j)]\ . \qquad (3)
\end{aligned}$$

In the above expression the indices i,j are summed over 1,2,3. The coefficients A, u_1, u_2, u_3, and u_4 are functions of the chemical potential and the temperature.

The stage has now been reached where both a microscopic expansion of the grand potential [Eq. (2)], and one based on symmetry arguments [Eq. (3)] have been obtained. Comparison of the two expansions gives microscopic expressions for the phenomenological coefficients. Explicit evaluation of the coefficients A, u_1, u_2, u_3, and u_4 in terms of a general band structure is carried out in the Appendix. Before proceeding to a discussion of the stability of various states (Sec. III) the grand potential expansion is transformed to a free-energy expansion.

E. Transformation to the free energy; Fermi-level shift (cf. Malaspinas and Rice, Ref. 16)

In order to describe the transition at a fixed number of particles, the free energy $F(T,N)$ is derived from $\Omega_t(T,\mu)$. As the temperature is lowered through the transition temperature the structure of the energy bands is changed, so that with a finite reservoir there is an accompanying shift in the Fermi level.

Denoting by h' the separation in energy of the crossing of the bands and the chemical potential μ (so that $\mu = -h' + \text{const}$), the trial number of particles is found from $N_{\text{trial}} = \partial\Omega_t(h')/\partial h'$. By differentiating

$$\Omega_t(h') = \Omega_0(h') + A(h') \sum_i (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{i}}) + \dots ,$$

one obtains

$$N_{\text{AF}}(h') - N_{\text{para}}(h') = \frac{\partial A(h')}{\partial h'} \sum_i (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{i}})$$

correct to second order in M^2, where N_{AF} and N_{para} denote the numbers of particles in the antiferromagnetic and paramagnetic phases, respectively. Setting $N_{\text{AF}}(h') = N_{\text{para}}(h)$ and denoting the total density of states for spins in one direction by N_T, then to order M^2,

$$2N_T(h - h') = \left[\frac{\partial A}{\partial h} \right] \sum_i (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{i}}) . \tag{4}$$

The above equation gives $h(h')$. Use can now be made of the theorem that the change in the free energy at fixed T,N equals the change in the grand potential at fixed T,μ, where the change is in some other independent quantity describing the state of the system (here M^2). Since the number of particles is a function only of h, and the chemical potential a function only of h', it sufficies to express $\delta\Omega_t$ in terms of h in order to obtain δF_t. Expressing $A(h')$ as $A(h) + (h'-h)\partial A/\partial h + \cdots$, and (since one is interested in an expansion to order M^4) neglecting higher-order terms, the free energy receives a correction

$$\left[\frac{1}{2N_T} \right] \left[\frac{\partial A}{\partial h} \right]^2 \left[\sum_i (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{i}}) \right]^2 = \left[\frac{1}{2N_T} \right] \left[\frac{\partial A}{\partial h} \right]^2 \left[\sum_\mu (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{i}}) + 2 \sum_{i>j} (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{i}})(\vec{\mathbf{M}}_j \cdot \vec{\mathbf{M}}_j) \right] , \tag{5}$$

so that for $F(T,N)$ the same coefficients result as for $\Omega(T,\mu)$, except that

$$u_1 \rightarrow u_1' = u_1 + \left[\frac{1}{2N_T} \right] \left[\frac{\partial A}{\partial h} \right]^2$$

and

$$u_3 \rightarrow u_3' = u_3 + \left[\frac{2}{2N_T} \right] \left[\frac{\partial A}{\partial h} \right]^2 . \tag{6}$$

The coefficients can now all be evaluated at $h' = h$.

III. THE COMPETITION OF MAGNETIZED STATES

A. Introduction

The model developed in the last section to describe the SDW state in chromium has led to a Landau free energy of the form $(i,j = 1,2,3)$

$$\begin{aligned} F = F + A \sum_i (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{i}}) + u_1' \sum_i (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{i}})^2 + u_2 \sum_i (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_i)(\vec{\mathbf{M}}_{\bar{i}} \cdot \vec{\mathbf{M}}_{\bar{i}}) + u_3' \sum_{i<j} (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_i)(\vec{\mathbf{M}}_{\bar{j}} \cdot \vec{\mathbf{M}}_{\bar{j}}) \\ + u_4 \sum_{i<j} [(\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_j)(\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{j}}) + (\vec{\mathbf{M}}_i \cdot \vec{\mathbf{M}}_{\bar{j}})(\vec{\mathbf{M}}_{\bar{i}} \cdot \vec{\mathbf{M}}_j)] . \end{aligned} \tag{7}$$

Among the various wave-vector and polarization states, the state realized by the system is the one which minimizes this free energy. Which state minimizes the free energy depends on the relative magnitudes of u_1, u_2, u_3, and u_4. In this chapter the physical nature of the realized state is determined for various relative u_1, u_2, u_3, and u_4.

Given the existence of the main component of the SDW, Eq. (7) is the simplest form for the free energy allowed by symmetry. As discussed in Sec.

(I), the ordered state in chromium contains higher harmonics of the fundamental SDW, as well as other fields, and these must be included in a more accurate Landau theory (see, e.g., Walker[27]). Here, however, the physical picture of a possible state is simply that of a magnetization density $\vec{M}(\vec{r})$ expanded in several Fourier waves, i.e.,

$$\vec{M}(\vec{r})=\sum_{\pm\vec{q}_i\ (i=1,2,3)} e^{i\vec{q}_i\cdot\vec{r}}\vec{M}_{q_i} .$$

For example, the single-$\vec{Q}$ state, linearly polarized in the x direction, i.e.,

$$\vec{M}(\vec{r})=2\hat{i}M_0\cos(\vec{q}\cdot\vec{r}+\phi_{qx}) ,$$

is obtained by choosing $|\vec{M}_{qx}|=M_0$, $(\vec{M}_q)_y=(\vec{M}_q)_z=0$. For a circularly polarized wave, choose $|M_{qx}|=|M_{qy}|=M_0$, $\phi_{qx}=\phi_q$, $\phi_{qy}=\phi_q-\pi/2$, so that

$$\vec{M}(\vec{r})=2\vec{M}_0[\hat{i}\cos(\vec{q}\cdot\vec{r}+\phi_q)\\ +\hat{j}\sin(\vec{q}\cdot\vec{r}+\phi_q)] .$$

Also, the possible simultaneous existence of two, or three waves is allowed for.

As there is no spin-orbit coupling in the model, the direction of polarization is undetermined. However, an anisotropic band structure does determine the direction of $\vec{q}$ relative to the lattice.

B. Minimization of the free energy

Denoting $(\vec{M}_{q_i})_\alpha$ by $|M_{i\alpha}|e^{i\phi_{i\alpha}}$, and noting that reality of $\vec{M}(\vec{r})$ requires $\vec{M}^*_{-q_i}=\vec{M}_{-q_i}$, then the second- and fourth-order terms in the free energy may be written

$$V_0=A\sum_i(|M_{ix}|^2+|M_{iy}|^2+|M_{iz}|^2) ,$$

$$V_1=u_1'\sum_i(|M_{ix}|^2+|M_{iy}|^2+|M_{iz}|^2)^2 ,$$

$$V_2=u_2\sum_i\left|\,|M_{ix}|^2e^{2i\phi_{ix}}+|M_{iy}|^2e^{2i\phi_{iy}}+|M_{iz}|^2e^{2i\phi_{iz}}\right|^2 ,$$

$$V_3=u_3'\sum_{i<j}(|M_{ix}|^2+|M_{iy}|^2+|M_{iz}|^2)(|M_{jx}|^2+|M_{jy}|^2+|M_{jz}|^2) ,$$

$$V_4=u_4\sum_{i<j}\Big(\left|\,|M_{ix}M_{jx}|e^{i(\phi_{ix}+\phi_{jx})}+|M_{iy}M_{jy}|e^{i(\phi_{iy}+\phi_{jy})}+|M_{iz}M_{jz}|e^{i(\phi_{iz}+\phi_{jz})}\right|^2\\ +\left|\,|M_{ix}M_{jx}|e^{i(\phi_{ix}-\phi_{jx})}+|M_{iy}M_{jx}|e^{i(\phi_{iy}-\phi_{jy})}+|M_{iz}M_{jz}|e^{i(\phi_{iz}-\phi_{jz})}\right|^2\Big) .$$

Since u_2,u_4 determine the polarization, it is convenient to consider four categories:

(1) $u_2,u_4>0$,

(2) $u_2,u_4<0$,

(3) $u_2<0$, $u_4>0$,

(4) $u_2>0$, $u_4<0$.

Cases (2) and (3) are the cases of interest for the models considered in the following; these are the only ones which will be discussed further.

(2) $u_2,u_4<0$. V_2 is made most negative by choosing the waves to be linearly polarized; each wave has the same polarization in order that V_4 contribute the largest negative term possible.

(3) $u_2<0$, $u_4>0$. The non-negative term V_4 is made to vanish by allowing no two waves to have polarization components in the same direction, V_2 is made most negative with linearly polarized waves.

Now attention is directed to finding the number of waves in the states. Considering again cases (2) and (3):

(2) Setting $A_i=|M_{ix}|^2$, $|M_{iz}|^2=|M_{iy}|^2=0$, then the terms in F are

$$V_0=A\sum_i A_i ,$$

$$V_1=u_1'\sum_i A_i^2 ,$$

$$V_2=u_2\sum_i A_i^2 ,$$

$$V_3=u_3'\sum_{i<j}A_iA_j ,$$

$$V_4=2u_4\sum_{i<j}A_iA_j .$$

The free energy is

$$F=F_0+A\sum_i A_i+(u_1'+u_2)\sum_i A_i^2+(u_3'+2u_4)\sum_{i<j}A_iA_j\ .$$

For the present it will be assumed that if two or more of the A_i are nonzero, then they have the same magnitude. In order that F be bounded from below for all values of the A_i, one must have $u_1'>0$ and $u_1'+u_2+u_3'+2u_4>0$. Depending on the relative magnitudes of $u_1'+u_2$ and $u_3'+u_4$, either the triple-$\vec{Q}$ or the single-$\vec{Q}$ state has lower free energy. One has for linearly polarized waves

(a) $u_3'+2u_4<2(u_1'+u_2)$ (triple-$\vec{Q}$) ,

(b) $u_3'+2u_4>2(u_1'+u_2)$ (single-$\vec{Q}$) .

(3) Let $A_1+|M_{1x}|$, $A_2=|M_{2y}|$, $A_3=|M_{3z}|$. Then

$$F=F_0+A\sum_i A_i+(u_1'+u_2)\sum_i A_i^2+u_3'\sum_{i<j}A_iA_j\ .$$

The regions of triple-$\vec{Q}$ and single-$\vec{Q}$ stability are, for linearly polarized waves

(a) $u_3'<2(u_1'+u_2)$ (triple-$\vec{Q}$) ,

(b) $u_3'>2(u_1'+u_2)$ (single-$\vec{Q}$) .

IV. EVALUATION OF THE FREE-ENERGY COEFFICIENTS FOR SPECIFIC MODELS

The purpose of this section is to evaluate the coefficients C_i of the Appendix in terms of several band structures, then to obtain the free-energy coefficients A, u_1, u_2, u_3, and u_4 which are given in terms of the C_i by Eqs. (A6), and finally, using the results of Sec. III, to draw conclusions regarding the relative stability of the various SDW states for each of the band structures.

A. Models for the band structure

The model Fermi surface considered to present that of chromium in what follows consists of two portions of similar shape (shown spherical below) separated in reciprocal space by half of a reciprocal-lattice vector. It is convenient to relabel the states of band b: $|bk\rangle\rightarrow|bk-G/2\rangle$, as shown in Fig. 1 below.

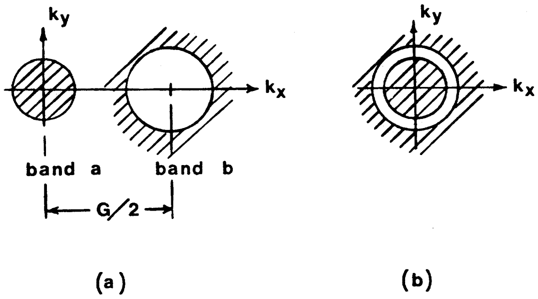

FIG. 1. Relabeling of band b states: (a) before relabeling, (b) after relabeling.

The two most widely used three-dimensional model band structures to represent that of chromium are the unequal sphere model (Rice[15]), and the octahedron model (Shibatani *et al.*[17]), shown in Fig. 2 below.

In the following the exact band structure is left unspecified. However, the concept of an energy h, measuring the deviation from perfect nesting of band a and band b, is retained. The energy of band α $(\alpha=a,b)$ may be written in the form

$$\epsilon_k^\alpha=h+\vec{v}_k^\alpha\cdot[\vec{k}-\vec{k}_F(\vec{k})]\ ,$$

where $\vec{k}_F(\vec{k})$ takes points $\vec{k}$ onto the nearest part of the Fermi surface directly above or below $\vec{k}$. Possible ambiguity in $\vec{k}_F(\vec{k})$ for points $\vec{k}$ far away from the Fermi surface is disregarded, as only states near the Fermi surface are important.

B. Expansion about the triple point

Referring to the free-energy expansion

$$F=A\sum_i(\vec{M}_i\cdot\vec{M}_{\bar{i}})+u_{1_i}'(\vec{M}_i\cdot\vec{M}_{\bar{i}})^2+\dots\ ,$$

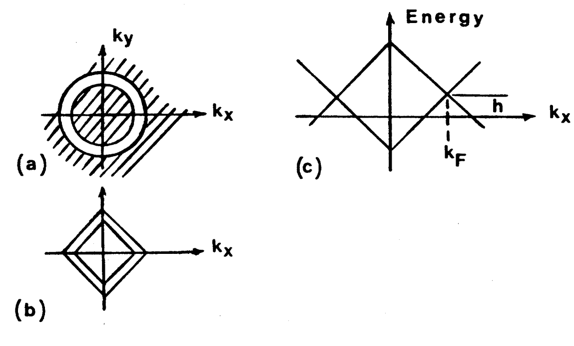

FIG. 2. Band structure for (a) the unequal sphere model and (b) the octahedron model.

the transition, assumed to be second order, occurs when $A(h,q,T)=0$. The wave vector $\vec{q}$ at the transition is the value which minimizes $A(h,q,T)$. For h less than a critical value h_0, this minimum occurs at $q^2=0$. At larger values of h, $q^2_{\min}$ increases continuously from zero. Nakanishi and Maki[18] noted that the free-energy coefficients could be evaluated analytically in the neighborhood of the point (h_0,T_0), the triple point. Here also, the evaluation of the coefficients, as a power series in q^2, is restricted to the neighborhood of the triple point.

C. General expansion

The calculation of the coefficients is accomplished most easily by directly expanding the propagator in a power series in q. Thus, expanding $\epsilon^{\alpha}_{k+q}=\epsilon^{\alpha}_{k}+\vec{v}^{\alpha}_{k}\cdot\vec{q}$, where $\vec{v}_k$ is the velocity of an electron of band $\alpha=a,b$, and defining $z_1=z_\nu-\epsilon^a_k$, $z_2-z_\nu-\epsilon^b_k$, one obtains the required coefficient C_0 [recall that $z_\nu=(2\nu+1)\pi i/\beta$],

$$\begin{aligned}C_0&=\frac{1}{2\beta}\sum_{k\nu}G^a_{k+q}G^b_k+(a\leftrightarrow b)\\&=\frac{1}{2\beta}\sum_{k\nu}\frac{1}{z_1z_2}\left[1+\frac{\vec{v}^a_k\cdot\vec{q}}{z_1}+\frac{(\vec{v}^a_k\cdot\vec{q})^2}{z_1^2}+\frac{(\vec{v}^a_k\cdot\vec{q})^3}{z_1^3}+\frac{(\vec{v}^a_k\cdot\vec{q})^4}{z_1^4}+\ldots+(a\leftrightarrow b)\right].\end{aligned}$$

Terms odd in $(\vec{v}_k\cdot\vec{q})$ vanish since $\vec{v}_{-k}=-\vec{v}_{-k}$ and $\epsilon^{\alpha}_{k}=\epsilon^{\alpha}_{-k}$, so that to order $(\vec{v}_k\cdot\vec{q})^4$,

$$C_0=\frac{1}{2\beta}\sum_{k\nu}\frac{1}{z_1z_2}\left[1+\frac{(\vec{v}^a_k\cdot\vec{q})^2}{z_1^2}+\frac{(\vec{v}^a_k\cdot\vec{q})^4}{z_1^4}+(a\leftrightarrow b)\right].$$

similarly, to order $(\vec{v}_k\cdot\vec{q})^2$,

$$C_1=\frac{1}{2\beta}\sum_{k\nu}\frac{1}{z_1^2z_2^2}\left[1+\frac{3(\vec{v}^a_k\cdot\vec{q})^2}{z_1^2}+(a\leftrightarrow b)\right],$$

$$C_2+C_3=\frac{1}{\beta}\sum_{k\nu}\frac{1}{z_1^2z_2^2}\left[1+\frac{(\vec{v}^a_k\cdot\vec{q})^2}{z_1^2}+(a\leftrightarrow b)\right],$$

$$\begin{aligned}C_4+C_6=\frac{1}{\beta}\sum_{k\nu}\frac{1}{z_1^2z_2^2}\Bigg[&1+\frac{1}{z_1^2}[4(\vec{v}^a_k\cdot\vec{q}_i)^2+2(\vec{v}^a_k\cdot\vec{q}_i)(\vec{v}^a_k\cdot\vec{q}_j)+(\vec{v}^a_k\cdot\vec{q}_j)^2]\\&+\frac{2}{z_1z_2}[(\vec{v}^a_k\cdot\vec{q}_i)(\vec{v}^b_k\cdot\vec{q}_i)+(\vec{v}^a_k\cdot\vec{q}_i)(\vec{v}^b_k\cdot\vec{q}_j)]+(a\leftrightarrow b)\Bigg],\end{aligned}$$

and

$$\begin{aligned}C_5=\frac{1}{2\beta}\sum_{k\nu}\Bigg[&\frac{1}{z_1^2z_2^2}1+\frac{1}{z_1^2}[2(\vec{v}^a_k\cdot\vec{q}_i)^2+2(\vec{v}^a_k\cdot\vec{q}_j)^2+3(\vec{v}^a_k\cdot\vec{q}_i)(\vec{v}^a_k\cdot\vec{q}_j)]\\&\frac{1}{z_1z_2}[(\vec{v}^a_k\cdot\vec{q}_i)(\vec{v}^b_k\cdot\vec{q}_j)+(\vec{v}^a_k\cdot\vec{q}_j)(\vec{v}^b_k\cdot\vec{q}_j)+(\vec{v}^a_k\cdot\vec{q}_i)(\vec{v}^b_k\cdot\vec{q}_i)+(\vec{v}^a_k\cdot\vec{q}_j)(\vec{v}^b_k\cdot\vec{q}_i)]+(a\leftrightarrow b)\Bigg].\end{aligned}$$

D. Specialization to simple Fermi surfaces

Both the unequal sphere model and the octahedron model have the property that a term such as $\sum_{k\nu}(1/z_1^2z_2^4)(\vec{v}_k\cdot\vec{q})^2$ can be written as the product of $\sum_{k\nu}(1/z_1^2z_2^4)$ and $\langle(\vec{v}_k\cdot\vec{q})^2\rangle$, where $\langle\ \rangle$ denotes an average over the Fermi surface. The restriction to such Fermi surfaces is now made. It is also assumed that $\vec{v}_k$ has a constant magnitude v over the Fermi surface. Since the Fermi surface has cubic symmetry, $\langle(\vec{v}_k\cdot\vec{q})^2\rangle$

$=\frac{1}{3}v^2q^2$. Thus, introducing the notation

$$\{rs\}=\frac{1}{\beta}\sum_{k\nu}\frac{1}{z_1^r z_2^s}\,,$$

$$C_0=\{11\}+\tfrac{1}{3}v^2q^2\{13\}+\langle(\vec{\mathrm{v}}_k\cdot\vec{\mathrm{q}})^4\rangle\{15\}\,,$$

$$C_1=\{22\}+v^2q^2\{24\}\,,$$

$$C_2+C_3=2\{22\}+\tfrac{2}{3}v^2q^2\{24\}\,,$$

$$C_4+C_6=2\{22\}+\tfrac{10}{3}v^2q^2\{24\}-\tfrac{4}{3}v^2q^2\{33\}\,,$$

$$C_5=\{22\}+\tfrac{4}{3}v^2q^2\{24\}-\tfrac{2}{3}v^2q^2\{33\}\,.$$

E. Evaluation of $\sum_k$

In evaluating, e.g.,

$$\{22\}=\frac{1}{\beta}\sum_{k\nu}\frac{1}{z_\nu-(h+\epsilon_k)}\,\frac{1}{z_\nu-(h-\epsilon_k)}\,,$$

the replacement $\sum_k\to N_m(0)\int_{-\infty}^{+\infty}d\epsilon$ is made, where $N_m(0)$ is the density of states of one of the bands. A consequence of this approximation is that the temperature dependence can be factored out, i.e.,

$$\{22\}=\frac{1}{\beta}N_m(0)\left[\frac{\beta}{\pi}\right]\times\left[\sum_\nu\int d\epsilon'\,\frac{1}{(2\nu+1)i-(\tilde{h}+\epsilon')}\times\frac{1}{(2\nu+1)i-(\tilde{h}-\epsilon')}\right],$$

where $\tilde{h}=(\beta/\pi)h$, so that the coefficients $u_i=u_i(h/T,q/T)$ $(i=1,\ldots,4)$. The second-order coefficient A, however, is still a function of h,q,T and not just of the scaled variables $h/T,q/T$. The reason for this is that $\{11\}$ formally diverges, showing that the approximation is inappropriate here.

An explicit formula for $\{rs\}$ has been worked out. It is

$$\{rs\}=2^{-s-r+3}\frac{(s+r-2)!}{(s-1)!(r-1)!}N_m(0)(\pi/\beta)^{s+r-2}\times\mathrm{Re}\left[i^{-s-r}\sum_{\nu\geq0}\frac{1}{(\tilde{\omega}_\nu+i\tilde{h})^{s+r-1}}\right],\qquad(8)$$

where $\omega_\nu=(2\nu+1)\pi/\beta$. The r and s dependence of $\{rs\}$ enters the h-dependent part of $\{rs\}$ only in the form $r+s$, so that the functions $\{13\}$, $\{22\}$, $\{15\}$, $\{24\}$, and $\{33\}$ can be expressed in terms of just two quantities α,β^*:

$$\alpha=\frac{2}{3}\left[\frac{\pi}{\beta}\right]^3\sum_{\nu\geq0}\frac{\omega^3-3\omega h^2}{(\omega^2+h^2)^3}\,,\qquad(9)$$

$$\beta^*=-\frac{2}{5}\left[\frac{\pi}{\beta}\right]^5\sum_{\nu\geq0}\frac{\omega^5-10\omega^3h^2+5\omega h^4}{(\omega^2+h^2)^5}\,.\qquad(10)$$

Before expressing the free-energy coefficients in terms of α,β^*, it is convenient to introduce dimensionless variables. Defining $\tilde{F}=[F/N_m(0)](\beta/\pi)^2$, $\tilde{M}^2=(\beta/\pi)M^2$, $\tilde{q}=vq\beta/2\pi$, $\tilde{h}=h\beta/\pi$, $\tilde{A}=A/N_m(0)$, and $\tilde{u}_i=[u_i/N_m(0)](\pi/\beta)^2$, all the new quantities (denoted by a tilde) become intensive and dimensionless. Referring now to the Appendix, giving the free-energy coefficents A,U_i $(i=1,\ldots,4)$ in terms of the C_j, one obtains

$$\begin{aligned}\tilde{A}(\tilde{h},\tilde{q},T)&=1/(N_m\bar{v})+\{\tilde{1}1\}+\alpha\tilde{q}^2\\&\quad+5\beta^*\tilde{q}^4\langle(\hat{v}_k\cdot\hat{q})^4\rangle\,,\\\tilde{u}_1'&=3\alpha+10\beta^*\tilde{q}^2+fE^2\,,\\\tilde{u}_2&=\tfrac{3}{2}\alpha-\tfrac{5}{3}\beta^*\tilde{q}^2\,,\\u_3'&=6\alpha+20\beta^*\tilde{q}^2+2fE^2\,,\\\tilde{u}_4&=6\alpha+\tfrac{20}{3}\beta^*\tilde{q}^2\,,\end{aligned}\qquad(11)$$

where fE^2 is the Fermi-level shift term, with $f=N_m/2N_T$ and $E^2=(\partial\tilde{A}/\partial\tilde{h})^2$.

Assuming a second-order transition, the transition occurs, on lowering the temperature, when $A(h,q,T)$ changes from being positive for all q to being negative for one of the q (h is held constant). By setting $\partial A/\partial\tilde{q}^2=0$ one finds that the value of $\tilde{q}^2$ at which the transition occurs is

$$\tilde{q}^2=-\frac{\alpha}{10\beta\langle(\hat{v}_k\cdot\hat{q})^4\rangle}\,.\qquad(12)$$

(Here the ^ on v_k and q denote unit vectors.) Now, by definition, the triple point is that point (in the hT plane) where both $A=0$ and $\tilde{q}^2=0$, so that one condition determining this point is $\alpha(h,T)=0$. By computation $\alpha=0$ for $h\simeq0.61(\pi/\beta)$, and as is clear from its definition [Eq. (9)], α is negative at larger values of h. By computation β^* is positive at $h=0.61(\pi/\beta)$. Because of the Fermi-surface average $\langle(\hat{v}_k\cdot\hat{q})^4\rangle$, the value of $\tilde{A}$ may be varied by changing the direction of $\hat{q}$. Since below the transition the free energy is lower the more negative is $\tilde{A}$, then $\hat{q}$ must be chosen so as to minimize $\langle(\hat{v}_k\cdot\hat{q})^4\rangle$.

F. Predictions of the models

Since the Fermi surface is invariant under transformations of the cubic group, the quantity $\langle(\hat{v}_k\cdot\hat{q})^4\rangle$ must be expressible in terms of the invariants of this group which can be formed from the components of $\hat{q}$. Thus one sets

$$\langle(\hat{v}_k\cdot\hat{q})^4\rangle=a_4(\hat{q}\cdot\hat{q})^2+a_4'(\hat{q}_x^4+\hat{q}_y^4+\hat{q}_z^4)\ .$$

On evaluating a_4, a_4' one obtains

(a) $a_4=\frac{1}{5}$, $a_4'=0$ for the spherical Fermi surface ,

(b) $a_4=\frac{1}{3}$, $a_4'=-\frac{2}{9}$ for the octahedral Fermi surface ,

(c) $a_4=0$, $a_4'=\frac{1}{3}$ for the cubic Fermi surface .

Of course for the spherical Fermi surface one direction for $\hat{q}$ gives as low a value to $\langle(\hat{v}_k\cdot\hat{q})^4\rangle$ as any other direction, namely $\frac{1}{5}$. For the octahedral Fermi surface it is favorable to have $\hat{q}$ point along one of the axes (as is the case experimentally), in which case $\langle(\hat{v}_k\cdot\hat{q})^4\rangle=\frac{1}{9}$. For the cubic Fermi surface it would be preferable for $\hat{q}$ to point in one of the $\{111\}$ directions, and this not being the case experimentally is understood since the actual Fermi surface does not much resemble a cube.

Substituting the value of $\langle(\hat{v}_k\cdot\hat{q})^4\rangle$ for each Fermi-surface model into (12), the magnitudes of $\tilde{q}^2$ for the cubic, spherical, and octahedral model are, respectively, $-\frac{3}{10}(\alpha/\beta^*)$, $-\frac{1}{2}(\alpha/\beta^*)$, and $-\frac{9}{10}(\alpha/\beta^*)$. The values of the free-energy coefficients may now be tabulated (see Table I).

With this information conclusions on the stability of the various states can be drawn.

(1) The finite positive value of the Fermi-level shift term fE^2 in u_1' and u_3' causes the general stability conditions to be satisfied. Therefore, near the triple point, any model with a noninfinite reservoir predicts a second-order transition.

(2) U_2 being negative causes the states to be linearly polarized. [Cases (2) and (3) of Sec. III B are relevant.]

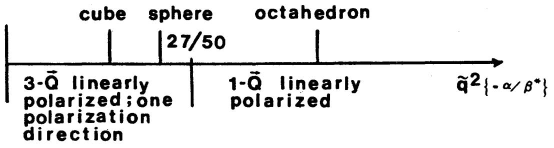

FIG. 3. Relative magnitudes of $\tilde{q}^2$ for the various Fermi-surface models.

(3) For the unequal sphere model, $u_2, u_4<0$ and $u_3'+2u_4<2(u_1'+u_2)$ so that the stable state is a triple-$\vec{Q}$ linearly polarized one, with all waves polarized in the same direction.

(4) For the octahedron model, $u_2<0$, $u_4=0$, and $u_3'+2u_4>2(u_1'+u_2)$ so that the single-$\vec{Q}$ linearly polarized state is stable.

(5) Assuming linearly polarized waves, with all waves polarized in the same direction ($u_2<0$, $u_4\geq 0$) then the condition $u_3'+2u_4>2u_1'+2u_2$ for a single-$\vec{Q}$ state requires [from Eqs. (11)] $\tilde{q}^2>-\frac{27}{50}\alpha/\beta^*$. This in turn implies that a value of $\langle(\hat{v}_k\cdot\hat{q})^4\rangle$ less than $\frac{5}{27}$ yields a single-$\vec{Q}$ state; if $\langle(\hat{v}_k\cdot\hat{q})^4\rangle$ is greater than $\frac{5}{27}$, then the triple-$\vec{Q}$ state is stable.

To summarize, the relative magnitudes of $\tilde{q}^2$ are illustrated above (see Fig. 3).

V. DISCUSSION

One of the results here is that the octahedron model predicts the single-$\vec{Q}$ linearly polarized state to be stable, whereas the sphere model predicts the triple-$\vec{Q}$ linearly polarized state to be stable. A qualitative explanation[28] for this comes from the nesting properties of the two models: In the unequal sphere model, translation of the hole sphere in, say the [100] direction, onto the electron sphere, and translation, say in the [010] direction, cause in the two cases different electron states to be paired, so that one might expect a triple-$\vec{Q}$ state to result. On the other hand, for the octahedron model the

TABLE I. Values of the free-energy coefficients for various Fermi-surface models.

Model of Fermi surface	u_1'	u_2	u_3'	u_4
Sphere	$-2\alpha+fE^2$	$\frac{7}{3}\alpha$	$-4\alpha+2fE^2$	$\frac{8}{3}\alpha$
Cube	fE^2	0	$2fE^2$	4α
Octahedron	$-6\alpha+fE^2$	3α	$-12\alpha+2fE^2$	0

above two translations cause some of the same electron states to be paired, so that there is some interference between the two waves (cf. Overhauser[11]).

With regard to previous results, Fenton[19] has also shown that the octahedron model predicts the single-$\vec{Q}$ state to be stable over the triple-$\vec{Q}$ state. He gives as criterion for the stability of the single-$\vec{Q}$ state that the Fermi velocity average perpendicular to the [111] direction in that octant must be much smaller than the parallel component. The present results support the general trend indicated by Fenton, but they also give a precise criterion for the stability of single-$\vec{Q}$ over multi-$\vec{Q}$ states. Nakanishi and Maki[18] find for the unequal sphere model that near the triple point the single-$\vec{Q}$ state is stable; they also state that near the impurity concentration where the transition temperature becomes 0 K, the triple-$\vec{Q}$ state appears most stable. The discrepany between the present results and those of Nakanishi and Maki is probably due to the latter authors' neglect of the finiteness of the electron reservoir.

Finally, a few remarks are in order on the occurrence of a first-order paramagnetic-incommensurate transition in chromium. The models considered in this paper give, within mean-field theory, second-order transitions near the triple point. The possibility that fluctuation effects may give rise to the appearance of a first-order transition is discussed by Bak, Krinsky, and Mukamel and by Fenton and Leavens. However, apart from such effects, the question remains as to what is the origin of the *actual* jump in the magnetization density. A mean-field mechanism for a first-order transition, with the harmonics playing an essential role, has been presented by Young and Sokoloff. This mechanism has the weakness, as noted by Fenton, that the three-band model is not in accord with any realistic band structure for chromium. Nonetheless, the possibility of a mean-field theory providing the explanation for the largest part of the first-order jump does not seem as remote to the present author as it does to some. In order to obtain such a jump near the triple point within the present theory, it would be necessary to introduce a nontrivial correlation between the separation in energy of the two bands (at points separated by the wave vector $\vec{Q}$) and the velocity of the electrons at these points. In this paper the final results have been simplified to the extent that the velocity of the electrons is constant over the Fermi surface.

APPENDIX: MICROSCOPIC DERIVATION OF THE GRAND POTENTIAL COEFFICIENTS

1. Diagram expansion of $\langle b_{k\sigma}^{\dagger} a_{k+q\sigma'} \rangle$

Given a Hamiltonian

$$H=\sum_i \epsilon_i c_i{}^{\dagger} c_i+\sum_{ij} \Delta_{ij} C_i{}^{\dagger} c_j \ ,$$

the thermal Green's function technique allows an expansion of

$$\langle c_j{}^{\dagger} c_i \rangle = Z^{-1}\,\mathrm{tr}(e^{-\beta H} c_j{}^{\dagger} c_i) \ ,$$

where $Z=\mathrm{tr}(e^{-\beta H})$, in the form

$$\langle c_j^{\dagger} c_i \rangle = \frac{1}{\beta} \sum_{\nu=-\infty}^{+\infty} e^{z_\nu 0^+} \left[\frac{1}{z_\nu - H} \right]_{ij} ,$$

where $z_\nu=(2\nu+1)\pi i/\beta$.

This expansion is now applied to the calculation of thermal averages with respect to the trial Hamiltonian $H_t=H_0+H_e$, where

$$H_e=\sum_{k,q} [a_{k+q}^{\dagger}(\vec{\mathrm{M}}_q\cdot\vec{\sigma})b_k+b_{k+q}^{\dagger}(\vec{\mathrm{M}}_q\cdot\vec{\sigma})a_k] \ .$$

Here the operators a_k,b_k are two-component spinor operators. This is to be understood when such operators are unaccompanied by a spin index σ,σ'. Iteration of the identity

$$\begin{aligned}(z_\nu-H_0-H_e)_{ij}^{-1} &= (z_\nu-H_0)_{ij}^{-1} \\ &\quad +((z_\nu-H_0)^{-1}H_e(z_\nu-H_0-H_e)^{-1})_{ij}\end{aligned}$$

provides the basis for a diagrammatic expansion of $\langle b_k^{\dagger} a_{k+q} \rangle$, which by definition represents the 2×2 matrix $(\langle b_{k\sigma}^{\dagger} a_{k+q\sigma'} \rangle)$. for example, the term first order in $\vec{\mathrm{M}}_q$ associated with $\langle b_k^{\dagger} a_{k+q} \rangle$ will be represented by the diagram shown in Fig. 4. As illustrated in the diagram, an external line $\vec{\mathrm{M}}_q\cdot\vec{\sigma}$ imparts a momentum q to an electron, changes its band (from a to b or from b to a), and multiplies the spinor by $(\vec{\mathrm{M}}_q\cdot\vec{\sigma})$. This expansion is now used to obtain an expansion of $\delta\Omega_t$ in powers of $\vec{\mathrm{M}}_q$.

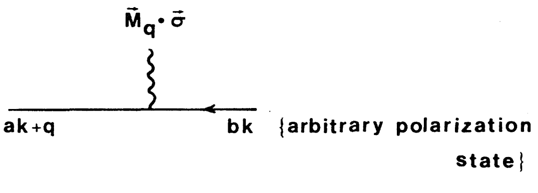

FIG. 4. First-order diagram for $\langle b_k^{\dagger} a_{k+q} \rangle$.

2. Diagrammatic expansion of $\delta\Omega_t$

Using the expansion of the thermal Green's function discussed in Sec. I the variation in the trial grand potential is expanded in a power series in $\{\vec{M}_q\}$.

a. Second-order part

Referring to Eq. (2) (Sec. II),

$$\delta\Omega_t^{(2)} = -\sum_{q_r\sigma\sigma} [(\vec{M}_q\cdot\vec{\sigma})_{\sigma\sigma'} + \bar{v}\psi^{(1)}_{\sigma\sigma'}(q)]\delta\psi^{(1)*}_{\sigma\sigma'}(q) + \text{c.c.} \tag{A1}$$

The first-order diagram for the propagator is as shown in Fig. 4, so that

$$\psi^{(1)}_{\sigma\sigma'}(q) = C_0(\vec{M}_q\cdot\vec{\sigma})_{\sigma\sigma'} ,$$

where

$$C_0 = \frac{1}{\beta}\sum_k G^a_{k+q}(z_\nu)G^b_k(z_\nu) .$$

Substituting into Eq. (A1)

$$\delta\Omega_t^{(2)} = -\sum_{q_r} C_0(1+\bar{v}C_0) \times \text{tr}[(\vec{M}_q\cdot\vec{\sigma})\sigma(\vec{M}_{-q}\cdot\vec{\sigma})] + \text{c.c.}$$

or

$$\delta\Omega_t^{(2)} = -4\sum_{q_r} C_0(1+\bar{v}C_0)(\vec{M}_q\cdot\delta\vec{M}_{-q}) .$$

Replacing $\sum_{q_r}$ by a sum over unordered pairs $\{q,-q\}$, one obtains

$$\Omega_t^{(2)} = -4\sum_{\text{pairs }\{q,-q\}} C_0(1+\bar{v}C_0)(\vec{M}_q\cdot\vec{M}_{-1}) . \tag{A2}$$

b. Fourth-order part

Referring again to Eq. (2),

$$\delta\Omega_t^{(4)} = -c_0\bar{v}\sum_{q_r\sigma\sigma'} \psi^{(3)}_{\sigma\sigma'}(q)\delta(\vec{M}_q\cdot\vec{\sigma})^*_{\sigma\sigma'} + \text{c.c.}$$

The second-order coefficient $1+\bar{v}c_0$ vanishes at the transition temperature so that $c_0\bar{v} = -1 + O(M^2)$. Therefore

$$\delta\Omega_t^{(4)} = \sum_{q_r\sigma\sigma'} [\psi^{(3)}_{\sigma\sigma'}(q)\delta(\vec{M}_q\cdot\vec{\sigma})^*_{\sigma\sigma'} + \text{c.c.}]$$

$$= \sum_{q_r} \text{tr}[\psi^{(3)}(q)(\delta\vec{M}_{-q})\cdot\vec{\sigma}] + \text{c.c.}$$

On substitution of

$$\psi_{\sigma\sigma'}(q) = \sum_{k\nu} e^{z_\nu 0^+} G_{ak+q\sigma,bk\sigma'}(z_\nu)$$

and dropping the factor $e^{z_\nu 0^+}$,

$$\delta\Omega_t^{(4)} = \frac{1}{\beta}\sum_{k\nu}\sum_{q_r} \text{tr}[G^{(3)}_{ak+q,bk}(z_\nu)\delta\vec{M}_{-q}\cdot\vec{\sigma}] + \text{c.c.} \tag{A3}$$

The propagator $G^{(3)}_{ak+q,bk}(z_\nu)$ consists of the sum of terms such as that shown in Fig. 5.

3. Evaluation of the general fourth-order coefficients

For counting purposes it is convenient to write the fourth-order part of the grand potential as

$$\Omega^{(4)} = \sum_{p,q,r,s} B(p,q;r,s)(\vec{M}_p\cdot\vec{M}_q)(\vec{M}_r\cdot\vec{M}_s) . \tag{A4}$$

It can be assumed that the coefficient $B(p,q;r,s)$ has the symmetry properties $B(p,q;r,s) = B(p,q;s,r) = B(r,s;p,q)$, that the q_i are ordered, and that $p \le q,r,s$ and $r \le s$. The terms can be classed according to the further ordering of p,q,r,s:

(a) Distinct wave vectors: $B(1234)$, $B(1324)$, $B(1423)$.

(b) One pair of wave vectors equal: $B(1123)$, $B(1233)$, $B(1322)$, $B(1213)$.

(c) Two pairs of wave vectors equal: $B(1122)$, $B(1212)$.

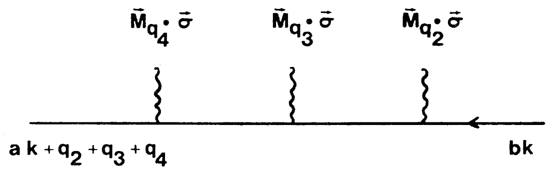

FIG. 5. A typical term in $G_{ak+q,bk}(z_\nu)$, where $\vec{q}_2+\vec{q}_3+\vec{q}_4=\vec{q}$.

The contribution of a given quadruple, say $\{p,q,r,s\}$, to $\delta\Omega_t^{(4)}$ is found by choosing one of the momenta, say p, and adding together all distinct diagrams arising from interchanges of the remaining three momenta.

By comparing terms in Eqs. (A3) and (A4), one obtains the following coefficients:

$$B(1,2;3,4)=\frac{2}{\beta}\sum_{k\nu}[G_2^aG_{234}^aG_{23}^bG_0^b-G_3^aG_{234}^aG_{23}^bG_0^b+G_3^aG_{234}^aG_{34}^bG_0^b+(3\leftrightarrow4)]\ ,$$

$$B(1,2;1,3)=\frac{2}{\beta}\sum_{k\nu}(G_1^aG_{113}^aG_{11}^bG_0^b+G_1^aG_{113}^aG_{13}^bG_0^b-G_3^aG_{113}^aG_{13}^bG_0^b)\ ,$$

$$B(1,1;2,3)=\frac{2}{\beta}\sum_{k\nu}(G_1^aG_{113}^aG_{11}^bG_0^b-G_1^aG_{113}^aG_{13}^bG_0^b+G_3^aG_{113}^aG_{13}^bG_0^b)\ ,$$

$$B(1,1;2,2)=\frac{2}{\beta}\sum_{k\nu}\tfrac{1}{2}(G_1^aG_{122}^aG_{12}^bG_0^b-G_2^aG_{122}^aG_{12}^bG_0^b+G_2^aG_{122}^aG_{22}^bG_0^b)\ ,$$

and

$$B(1,2;1,2)=\frac{2}{\beta}\sum_{k\nu}G_2^aG_{122}^aG_{12}^bG_0^b\ .$$

The notation above has, for example, $G_{122}=G_{k+q_1+q_2+q_2}$.

4. The Landau coefficients (specialized to Cr)

The coefficients A, u_1, u_2, u_3, and u_4 of Sec. II D can now be written in terms of the band structure.

a. Second-order coefficient

One obtains

$$A=-4C_0(1+\bar{v}C_0)\ ,$$

where

$$C_0=\frac{1}{\beta}\sum_{k\nu}G_{k+q}^a(z_\nu)G_k^b(z_\nu)\ .$$

b. Fourth-order coefficients

Setting $1=i$ and $2=\bar{i}$ in the equation for $B(1,2;1,2)$, one obtains

$$u_1=\frac{2}{\beta}\sum_{k\nu}(G_{k+q_i}^a)^2(G_k^b)^2=2C_1\ .$$

Also, from $B(1,1;2,2)$,

$$u_2=\frac{1}{\beta}\sum_{k\nu}[G_{k+q_i}^aG_{k-q_i}^a(G_k^b)^2-(G_{k+q_i}^a)^2(G_k^b)^2+(G_{k+q_i}^a)^2G_{k+2q_i}^bG_k^b]$$

or

$$u_2=C_2-C_1+C_3\ .$$

Next, evaluating the three cases of four distinct momenta, $\{\,i,\bar{i},j,\bar{j}\,\}$:

$$B(i,\bar{i};j,\bar{j},)=\frac{2}{\beta}\sum_{k\nu}[G^a_{k-q_i})^2G^b_{k-q_i+q_j}G^b_k-G^a_{k-q_j}G^a_{k-q_i}G^b_{k-q_i+q_j}G^b_k+G^a_{k+q_j}G^a_{k-q_j}(G^b_k)^2+(G^a_{k-q_i})^2G^b_{k-q_i-q_j}G^b_k$$

$$-G^a_{k-q_j}G^a_{k-q_i}G^b_{k-q_i-q_j}G^b_k+G^a_{k-q_j}G^a_{k-q_i}(G^b_k)^2]$$

or

$$B(i,\bar{i};j,\bar{j})=2(\bar{C}_4-\bar{C}_5+C_6+C_4-C_5+\bar{C}_6)\ .$$

Similarly,

$$B(i,j;\bar{i},\bar{j})=2(C_4-\bar{C}_4+C_5+\bar{C}_5+C_6-\bar{C}_6)$$

and

$$B(i,\bar{j};\bar{i},j)=2(-C_4+\bar{C}_4+C_5+\bar{C}_5-C_6+\bar{C}_6)\ ,$$

where

$$\begin{aligned}
C_1&=\frac{1}{\beta}\sum_{k\nu}(G^a_{k+q_i})^2(G^b_k)^2\ ,\\
C_2&=\frac{1}{\beta}\sum_{k\nu}(G^a_{k+q_i})(G^b_{k-q_i})(G^b_k)^2\ ,\\
C_3&=\frac{1}{\beta}\sum_{k\nu}(G^a_{k+q_i})^2(G^b_{k+2q_i})G^b_k\ ,\\
C_4&=\frac{1}{\beta}\sum_{k\nu}(G^a_{k+q_i})^2G^b_{k+q_i+q_j}G^b_k\ ,\\
\bar{C}_4&=\frac{1}{\beta}\sum_{k\nu}(G^a_{k+q_i})^2G^b_{k+q_i-q_j}G^b_k\ ,\\
C_5&=\frac{1}{\beta}\sum_{k\nu}G^a_{k+q_i}G^a_{k+q_i}G^b_{k+q_i+q_j}G^b_k\ ,\\
\bar{C}_5&=\frac{1}{\beta}\sum_{k\nu}G^a_{k+q_i}G^a_{k-q_j}G^b_{k+q_i-q_j}G^b_k\ ,\\
C_6&=\frac{1}{\beta}\sum_{k\nu}G^a_{k+q_i}G^a_{k-q_j}(G^b_k)^2\ ,\\
\bar{C}_6&=\frac{1}{\beta}\sum_{k\nu}G^a_{k+q_i}G^a_{k+q_j}(G^b_k)^2\ .
\end{aligned}\tag{A4}$$

From $u_3=B(i,\bar{i};j,\bar{j})$ and $u_4=B(i,j;\bar{i},\bar{j})$ one obtains

$$u_3=2(C_4+\bar{C}_4-C_5-\bar{C}_5+C_6+\bar{C}_6)$$

and

$$u_4=2(C_5+\bar{C}_5)\ .$$

Under band interchange ($a\leftrightarrow b$),

$$\begin{aligned}
C_2&=\frac{1}{\beta}\sum_{k\nu}(G^a_{k+q_i})(G^a_{k-q_i})(G^b_k)^2\\
&\rightarrow\frac{1}{\beta}\sum_{k\nu}(G^a_k)(G^b_{k+q_i})(G^b_{k-q_i})\\
&=\frac{1}{\beta}\sum_{k\nu}(G^a_{k+q_i})^2(G^b_{k+2q_i})(G^b_k)=C_3\ .
\end{aligned}$$

Similarly under band interchange, $C_4\rightarrow C_6$, $C_4\rightarrow C_6$, and $C_5\rightarrow\bar{C}_5$. Also, under reflection in an appropriate plane, $C_4\rightarrow C_4$, $C_5\rightarrow C_5$, and $C_6\rightarrow C_6$; since the energy bands are invariant under this reflection, one has $C_4=\bar{C}_4$, $C_5=\bar{C}_5$, and $C_6=\bar{C}_6$. Thus

$$\begin{aligned}
A&=-4C_0(1+\bar{v}C_0)\ ,\\
u_1&=2C_1\ ,\\
u_2&=C_2-\tfrac{1}{2}C_1+a\leftrightarrow b\ ,\\
u_3&=4C_4-2C_5+a\leftrightarrow b\ ,\\
u_4&=4C_5+a\leftrightarrow b\ .
\end{aligned}\tag{A6}$$

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