Elastic and magnetic interactions in a narrow twofold-degenerate band

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The interplay between the elastic and magnetic interactions in a twofold-degenerate band has been studied under the Hartree-Fock approximation. The free energy of the system has been expanded in terms of tetragonal-type elastic strain and magnetic moment and this is then used to study the possibility of the coexistence of ferromagnetic and tetragonally distorted phases and various anomalies in the physical properties of such a system. The experimental results in some real systems are discussed on the basis of the present analysis.

I. INTRODUCTION

There has been considerable interest in the last decade in understanding the interplay between the structural and superconducting transitions in $A15$ and other intermetallic compounds arising from narrow and degenerate d bands.[1] Since the interactions between the d electrons (Coulomb and exchange) responsible for the magnetic properties also play an important role in narrow-band systems, the study of the interplay between the electron-phonon interaction, particularly that of the elastic type which favors structural transitions of the tetragonal type in a cubic crystal, and the magnetic interactions arising from the same d electrons is of current interest.[2–6] There are two approaches in the study of this interplay:

(i) Detailed band calculations taking into account the change in the band energy due to the lattice distortion and incorporating at the same time the Coulomb and exchange interactions between the degenerate d electrons such as that recently done for fcc Mn by Oguchi and Freeman[3] and for bcc iron by Hasegawa *et al.*[4]

(ii) Model calculations based on degenerate Hubbard bands incorporating both elastic and magnetic interactions in a self-consistent way.[5,6]

The band calculations based on local-density-functional formalism[7] give good values of the ground-state energy—at least for systems where the Coulomb correlation between the d electrons is smaller than the bandwidth. But for calculating the temperature dependence of physical parameters such as the elastic constant, the magnetic susceptibility, and the specific heat and their anomalies near phase transition temperatures, the accuracy of band calculations is often questionable. In general, the finite-temperature properties in which the low-lying excited states take part still pose difficulties in band calculations. Also, the size of the numerical computations involved in the band calculations increases enormously when the number of atoms per unit cell is large. In our present problem of degenerate band systems, the electron-phonon interaction plays an important role and it is necessary to treat both magnetic and electron-phonon interactions in a self-consistent way. This is a rather difficult task from the band point of view. The model calculations, on the other hand, have both their usefulness and limitations. The choice of a model density of states (DOS) appropriate to the real systems is one of the principal difficulties. Another uncertainty arises in fixing the values of the parameters of the model. But this can be avoided by estimating these parameters from band calculations for the bare electrons. The model calculations have the advantage that different approximations involved in treating the many-body Hamiltonian for the d electrons can be systematically studied.[8] In many cases one gets results independent of the form of the DOS and the general features of a large class of compounds can be studied, whereas the band calculations have to be done for each compound separately. The finite-temperature effects can be studied within the model calculations through the Fermi function, incorporating the fluctuations of local spin and strains either through the functional integral method[9] or through appropriate averaging of the terms appearing in the Landau-Ginzburg expansion of the free energy.[10]

In the present paper we have treated the problem of the interplay between elastic and magnetic interactions in a twofold-degenerate band under the Hartree-Fock approximation in a self-consistent way and have studied the physical properties of such an electronic system. The case of a twofold-degenerate band is of particular interest. The Fermi energy (E_F) lies on a high DOS arising from e_g-type electrons for many of the transition metals and their compounds. The recent band calculations on bcc iron done by Hasegawa *et al.*[4] show that E_F lies close to the peak of an e_g-type DOS. The same is true for γ-Mn.[3,11] The analysis of experimental results of La_3S_4 and La_3Se_4 which undergo cubic to tetragonal transitions and where the magnetic interactions play an important role indicates the same situation.[12] The band calculations[13] and available experimental results on the TiH_{2-x} system also show similar behavior.[14]

II. MODEL HAMILTONIAN AND FREE-ENERGY EXPANSION

The coupling between the e_g electrons and the shear mode can arise either through the modification of the DOS due to the distortion of the lattice or through the

direct electron-phonon interaction between the e_g electrons and the same type of elastic mode. The first process would make the bandwidths of the two orbital subbands different. Ghatak *et al.*[15] have shown that the change in the free energy from the cubic to tetragonal phase does not significantly depend on this variation of bandwidth. On the contrary, the second process reproduces correctly the essential features of the structural transition. In this paper we, therefore, consider the second process and write the model Hamiltonian as

$$\mathcal{H}=\sum_{i,j,\sigma} t_{ij}(C^{\dagger}_{i1\sigma}C_{j1\sigma}+C^{\dagger}_{i2\sigma}C_{j2\sigma})+U\sum_{i}(n_{i1\uparrow}n_{i1\downarrow}+n_{i2\uparrow}n_{i2\downarrow})+U'\sum_{i,\sigma,\sigma'} n_{i1\sigma}n_{i2\sigma'}$$
$$-J\sum_{i,\sigma} n_{i1\sigma}n_{i2\sigma}+Ge\sum_{i,\sigma}(n_{i1\sigma}-n_{i2\sigma})+\tfrac{3}{4}NC_0e^2-\mu_B H\sum_{i}[(n_{i1\uparrow}-n_{i1\downarrow})+(n_{i2\uparrow}-n_{i2\downarrow})]\,, \tag{1}$$

where N is the number of atoms, 1 and 2 denote the orbitals, U and U' are intra-atomic intraorbital and interorbital Coulomb interaction terms, J is the exchange interaction between the two orbitals at the same site, G is the strength of the interaction between e_g-type electrons and strain modes, C_0 $(=C_{11}-C_{12})$ is the tetragonal-type elastic constant for the lattice arising from all other electrons except the e_g electrons, and the last term is the Zeeman term. The spin-dependent electron numbers for the two orbitals are denoted by $n_{i\gamma\sigma}$ where γ is the orbital index. We then made the Hartree-Fock approximation so that the product of two operators $A_\alpha A_\beta$ is written as

$$A_\alpha A_\beta=\langle A_\alpha\rangle A_\beta+\langle A_\beta\rangle A_\alpha-\langle A_\alpha\rangle\langle A_\beta\rangle\,, \tag{2}$$

where $\langle\ \rangle$ denotes the thermal average. The Fourier-transformed Hamiltonian in the Hartree-Fock approximation can then be expressed as

$$\mathcal{H}^{\rm HF}=\sum_{K,\gamma,\sigma} E_{K\gamma\sigma}n_{K\gamma\sigma}+\tfrac{3}{4}NC_0e^2$$
$$-NU(n_{1\uparrow}n_{1\downarrow}+n_{2\uparrow}n_{2\downarrow})$$
$$-NU'\sum_{\sigma,\sigma'} n_{1\sigma}n_{2\sigma'}+NJ\sum_{\sigma} n_{1\sigma}n_{2\sigma}\,, \tag{3}$$

where $\langle n_{i\gamma\sigma}\rangle=n_{i\gamma\sigma}=n_{\gamma\sigma}$. This is valid in the paramagnetic and ferromagnetic phases. The antiferromagnetic case would be treated separately. The energies of the four spin-orbit subbands $E_{K\gamma\sigma}$ are given by

$$E_{K\gamma\sigma}=\epsilon_K+\lambda\left[Ge+\frac{A}{4}\delta n\right]-\sigma\left[\frac{Bm}{4}+\mu_B H\right]+\sigma\lambda\frac{D}{4}\delta m\,, \tag{4}$$

where $\sigma=\pm1$ for the spin-up and spin-down, respectively, $\lambda=\pm1$ for the orbitals 1 and 2, and $A=2U'-U-J$, $B=U+J$, $D=U-J$. We have also defined $\delta n=n_2-n_1$, $m=n_\uparrow-n_\downarrow=(n_{1\uparrow}+n_{2\uparrow})-(n_{2\downarrow}+n_{2\downarrow})$, and $\delta m=(n_{2\uparrow}-n_{2\downarrow})-(n_{1\uparrow}-n_{1\downarrow})$ so that m gives the total magnetic moment per atom due to the two orbitals, whereas δm gives the difference in the moment in the two orbitals. This difference in the moment arises when the numbers of electrons in the two orbitals are different due to the strain e and can play a significant role in our problem. We have neglected a constant shift of band energy in the cubic-paramagnetic phase ϵ_K due to Coulomb and exchange interactions, e.g., $(E/4)n$ where $E=U+2U'-J$ and $n=\sum_{\gamma,\sigma} n_{\gamma\sigma}$, the total number of electrons per atom. The Hartree-Fock Hamiltonian can further be expressed as

$$\mathcal{H}^{\rm HF}=\sum_{k,\gamma,\sigma} E_{k\gamma\sigma}n_{k\gamma\sigma}+\tfrac{3}{4}NC_0e^2$$
$$+\frac{N}{8}A(\delta n)^2+\frac{N}{8}Bm^2+\frac{N}{8}D(\delta m)^2\,. \tag{5}$$

The free energy per atom corresponding to this Hamiltonian can then be written as

$$F=-\frac{1}{\beta}\frac{1}{N}\sum_{k,\gamma,\sigma}\ln\{1+\exp[-\beta(E_{k\gamma\sigma}-\mu)]\}$$
$$+\sum_{k,\gamma,\sigma}\mu n_{k\gamma\sigma}+\tfrac{3}{4}C_0e^2+\frac{A}{8}(\delta n)^2+\frac{B}{8}m^2+\frac{D}{8}(\delta m)^2\,, \tag{6}$$

where $\beta=1/k_BT$ and μ is the chemical potential in the transformed phase, given by $\mu=\mu_{\rm PC}+\Delta\mu$, $\mu_{\rm PC}$ being the chemical potential in the paramagnetic-cubic phase. This follows from the thermodynamic equilibrium of the electrons in the four subbands in the final transformed phase.

There are in all four order parameters, e, δn, m, and δm, implied in the expression of F given by Eq. (6). In our problem the two principal order parameters are e and m. It is necessary to express the two subsidiary order parameters δn and δm as functions of e and m and expand the free energy F in terms of these two order parameters at least up to fourth order to study the phase transitions. We have done this following the subsequent prescription:

$$\delta n=n_2-n_1=\sum_{k,\sigma}[f(E_{k2\sigma})-f(E_{k1\sigma})]\,, \tag{7}$$

where $f(E)$ is the Fermi function which is defined as

$$f(E)=\frac{1}{1+\exp[\beta(E-\mu)]} \tag{8}$$

and

$$\delta m=\sum_{k,\sigma}\{[f(E_{k2\uparrow})-f(E_{k2\downarrow})]$$
$$-[f(E_{k1\uparrow})-f(E_{k1\downarrow})]\}\,. \tag{9}$$

By expanding the Fermi functions about their values in the paramagnetic-cubic (PC) phase we express δn and δm as functions of e, m, $\Delta\mu$, and I_n, where

$$I_n=\int_{-\infty}^{\infty}\rho(\epsilon)\left[-\frac{\partial^n f}{\partial\epsilon^n}\right]d\epsilon \tag{10}$$

are the Stoner integrals.[16] $\rho(\epsilon)$ is the DOS per spin orbital per atom in the PC phase. The conservation of the number of electrons in the PC and FT (ferromagnetic-tetragonal) phase enables us to express $\Delta\mu$ in terms of other variables. We then arrive at the following expression for δn and δm where we have retained terms up to third powers of e, m, and field H which is sufficient to get the free energy up to fourth order in the order parameters

$$\delta n=ae+be^3+ce\left[\mu_B H+\frac{B}{4}m\right]^2 \tag{11}$$

and

$$\delta m=de\left[\mu_B H+\frac{B}{4}m\right], \tag{12}$$

where

$$\begin{aligned}
a&=\frac{4GI_1}{1-AI_1}\,,\\
b&=-\frac{2}{3}\frac{G^3I_1X_2}{(1-AI_1)^4}\,,\\
c&=-\frac{2GI_1X_3}{(1-AI_1)^2}\,,\\
d&=-\frac{4I_2G}{(I-DI_1)(1-AI_1)}\,,\\
X_2&=3\left[\frac{I_2}{I_1}\right]^2-\frac{I_3}{I_1}\,,\\
X_3&=\left[\frac{I_2}{I_1}\right]^2-\frac{I_3}{I_1}-\frac{2DI_1}{1-DI_1}\left[\frac{I_2}{I_1}\right]^2,
\end{aligned} \tag{13}$$

and the free energy is then given by

$$\begin{aligned}
F=F_{\rm PC}&+\tfrac{1}{2}a_2e^2+\tfrac{1}{4}a_4e^4+\tfrac{1}{2}b_2m^2+\tfrac{1}{4}b_4m^4+\tfrac{1}{2}Ce^2m^2\\
&+\eta_1\mu_B Hm+\tfrac{1}{2}\eta_2(\mu_B H)^2+\tfrac{1}{4}\eta_3e^2(\mu_B H)^2\\
&+\eta_4e^2\mu_B Hm\,,
\end{aligned} \tag{14}$$

where

$$\begin{aligned}
a_2&=\frac{[1-(A+4\alpha)I_1]G^2}{\alpha(1-AI_1)}\,,\\
a_4&=\frac{2}{3}\frac{I_1X_2G^4}{(1-AI_1)^4}\,,\\
b_2&=\frac{B}{4}(1-BI_1)\,,\\
b_4&=\frac{2}{3}\left[\frac{B}{4}\right]^4I_1X_2\,,\\
C&=2\left[\frac{B}{4}\right]^2\frac{I_1X_3G^2}{(1-AI_1)^2}\,,
\end{aligned} \tag{15}$$

$$\begin{aligned}
\eta_1&=-BI_1\,,\\
\eta_2&=-4I_1\,,\\
\eta_3&=\frac{4I_1G^2X_3}{(1-AI_1)^2}\,,\\
\eta_4&=\frac{BI_1G^2X_3}{2(1-AI_1)^2}\,,
\end{aligned}$$

with $\alpha=2G^2/3C_0$. The free energy expansion given by Eq. (14) is valid at finite temperature and for any form of DOS and is, therefore, more general than that given by Ghatak and Ray.[5] This is an extension of the Stoner-Wohlfarth theory[17] to the cases where elastic interactions are important.

It is evident from the expressions given by Eqs. (11) and (15) that the factor $1-AI_1$ plays an important role. Since $A=2U'-U-J$, this factor signifies the Coulomb- and exchange-induced transfer of electrons from one orbital to another and can considerably modify the elastic effects even in the absence of magnetic ordering. The interorbital Coulomb interaction U' favors elastic transitions, whereas the intraorbital Coulomb U and the interorbital exchange would oppose such transitions. It is to be noted that for the e_g-band case considered here, there are no odd-order terms in e in the free-energy expansion. Therefore both elastic and magnetic transitions are expected to be second order. But if the DOS, due to other types of electrons (e.g., t_{2g} or s and p) is not negligible—particularly near $E=E_F$—such an odd-order term in F would be nonvanishing and might change the order of the phase transition. Also, if E_F lies close to a minimum in the DOS, a_4 and b_4 might become negative and consequently the transition would be first order as is evident from the following expression for I_1:

$$I_1=\rho(\epsilon_f)\left\{1-\frac{\pi^2}{6}(k_BT)^2\left[\left[\frac{\rho'(\epsilon_F)}{\rho(\epsilon_F)}\right]^2-\frac{\rho''(\epsilon_F)}{\rho(\epsilon_F)}\right]\right\}, \tag{16}$$

where ρ' and ρ'' are derivatives of ρ with respect to ϵ at $\epsilon=E_F$. It can be shown that only if E_F lies near a minimum in the DOS, then $\rho'\simeq0$ and ρ'' is positive, and consequently the fourth-order terms a_4 and b_4 can become negative making the transitions first order. Such a situation can arise if the DOS has a two-peaked structure. But for all single-peaked DOS, the transitions would be second-order, and we shall consider such transitions here leaving the cases of first-order transitions to be treated separately.

Another point worth mentioning is that the main effects of δm appears through the parameter C in Eq. (14). The factor

$$\frac{2DI_1}{1-DI_1}\left[\frac{I_2}{I_1}\right]^2$$

in C is due to δm. This can become large since near the magnetic transition where $BI_1\simeq1$, the value of DI_1 would be comparable to 1 except for the half-filled-band case where $I_2\simeq\rho'(\epsilon_F)=0$. Also, for the e_g band $v'=v-2J$.

The order parameters e and m for PC→PT (PT represents paramagnetic tetragonal) and PC→FC (FC represents ferromagnetic-cubic) transitions are obtained by minimizing the free energy with respect to e and m, respectively, and are given by

$$e^2=-\frac{a_2}{a_4}=\frac{3}{2G^2}\frac{[(A+4\alpha)I_1-1](1-AI_1)^3}{\alpha I_1 X_2}, \quad (17)$$

$$m^2=-\frac{b_2}{b_4}=\frac{3}{2}\left[\frac{4}{B}\right]^3\frac{(BI_1-1)}{I_1X_2}. \quad (18)$$

The temperature T_M is obtained from Eq. (17) as

$$(K_BT_M)^2=\frac{6}{\pi^2}\frac{[(A+4\alpha)\rho(\epsilon_F)-1]}{(A+4\alpha)\rho(\epsilon_F)X_1}, \quad (19)$$

where

$$X_1=\left[\frac{\rho'(\epsilon_F)}{\rho(\epsilon_F)}\right]^2-\frac{\rho''(\epsilon_F)}{\rho(\epsilon_F)}. \quad (20)$$

From Eqs. (17) and (19) we get

$$e=\begin{cases}P\left[1-\dfrac{T^2}{T_M^2}\right]^{1/2} & \text{for } T<T_M,\\ 0 \text{ otherwise,}\end{cases} \quad (21)$$

where

$$P=\left[\frac{\pi^2}{4\alpha G^2}(A+4\alpha)\frac{X_1}{X_2}[1-A\rho(\epsilon_F)]^3\right]^{1/2}k_BT_M.$$

For T close to T_M, the HF theory gives

$$e=\sqrt{2}P\left[1-\frac{T}{T_M}\right]^{1/2}. \quad (22)$$

Similarly, Eq. (18) gives the Curie temperature T_C as

$$(k_BT_C)^2=\frac{6}{\pi^2}\frac{B\rho(\epsilon_F)-1}{B\rho(\epsilon_F)X_1}. \quad (23)$$

We consequently obtain

$$m=\begin{cases}Q\left[1-\dfrac{T^2}{T_C^2}\right]^{1/2} & \text{for } T<T_C,\\ 0 \text{ otherwise,}\end{cases} \quad (24)$$

with

$$Q=\left[\pi^2\left[\frac{4}{B}\right]^2\frac{X_1}{X_2}\right]^{1/2}k_BT_C.$$

The critical exponent for the two order parameters e and m is, therefore, 0.5 in this mean-field theory. The recent measurements of $e=c/a-1$ in La_3S_4 by Westerholt *et al.*[12] show that for T very close to T_M, $\beta=0.31$ indicating that fluctuations of local strains play an important role close to the transition temperature.

It is evident from Eq. (17) that the magnitude of e might be modified by the Coulomb and exchange interactions through the parameter A. The magnetic moment in the FC phase is not affected by the uniform strain e but the local fluctuations in the value of e might alter the magnitude of m and T_C in degenerate band systems.

III. PHASE DIAGRAM AT $T=0$

On the basis of the free energy given by Eq. (15) the phase diagram at $T=0$ can be studied for any model DOS and for any band filling. The results depend on the form of the DOS through the derivatives of ρ at $\epsilon=\epsilon_F$. The interesting case is that of the half-filled band where $\rho'(\epsilon_F)=0$ and consequently the energies in different phases can be expressed as multiples of I_3/I_1 $[=\rho''(\epsilon_F)/\rho(\epsilon_F)]$. The relative stability of different phases can then be compared.

In Fig. 1 we give the phase diagram for this case for three different values of A. For $A=0$, the diagram is symmetric between the PT and FC phases, whereas for $A>0$ the region of the PT phase increases and the contrary is true for $A>0$.

The ferromagnetic-tetragonal phase is not stable for this case. This is in conformity with the previous results obtained by Ghatak and Ray for the parabolic DOS, where the coexistence of the ferromagnetism and the tetragonal distortion was found not to be possible for any filling of the band. Since the coupling parameter C in Eq. (18) is positive with a DOS having a single peak (and consequently without any minimum), the energy always increases for simultaneous existence of m and e, both of which have the same wave vector $\mathbf{K}=0$ and have infinite wavelength. It would be interesting to see whether a first-order ferromagnetic-tetragonal transition is possible when E_F lies in a minimum of a double-peaked DOS.

IV. MAGNETIC SUSCEPTIBILITY IN THE PARAMAGNETIC PHASE

The experimental results for the magnetic susceptibility χ in La_3S_4, La_3Se_4, and TiH_2 clearly show the existence of a peak at $T=T_M$, the tetragonal transition temperature. Also, $\partial\chi/\partial T$ changes sign at $T=T_M$ with

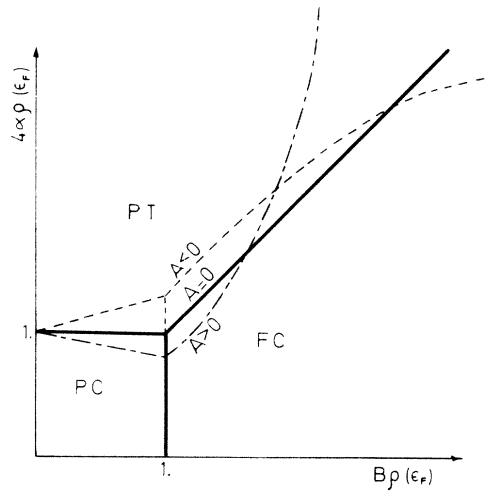

FIG. 1. Phase diagram for the half-filled-band case.

$$\left|\left[\frac{\partial\chi}{\partial T}\right]_{T>T_M}\right| < \left|\left[\frac{\partial\chi}{\partial T}\right]_{T<T_M}\right| .$$

In order to explain these results the following equation of state is obtained from the condition $\partial F/\partial m=0$:

$$(b_2+Ce^2)m+b_4m^3=-\mu_B H(\eta_1+\eta_4 e^2) . \tag{25}$$

The derivative of this equation with respect to H then gives the magnetic susceptibility as

$$\chi(T)=\frac{4\mu_B^2 I_1\left[1-\frac{1}{2}\frac{(Ge)^2X_3}{(1-AI_1)^2}\Theta(T_M-T)\right]}{1-BI_1\left[1-\frac{1}{2}\frac{(Ge)^2X_3}{(1-AI_1)^2}\Theta(T_M-T)\right]} , \tag{26}$$

where

$$\Theta(T_M-T)=\begin{cases}1 & \text{for } T<T_M\\ 0 & \text{othewise.}\end{cases}$$

This expression clearly shows that $\chi(T)$ increases as temperature is lowered from the high-temperature side in the PC phase since I_1 increases with a decrease of temperature and reaches a maximum at $T=T_M$. Below T_M, e increases, and consequently χ decreases. Substituting the expression of e^2 from Eq. (17) in Eq. (26) and making expansion of $\chi(T)$ about $T=T_M$, we finally get

$$\frac{\left[\frac{\partial\chi}{\partial T}\right]_{T<T_M}}{\left[\frac{\partial\chi}{\partial T}\right]_{T>T_M}}=1-\Gamma , \tag{27}$$

where $\Gamma=3(X_3/X_2)$. For the half-filled-band case $X_3=X_2$ and, consequently, we get

$$\left|\frac{\chi'(T<T_M)}{\chi'(T>T_M)}\right|=2 . \tag{28}$$

We give a plot of $\chi(T)$ versus T in Fig. 2 for the half-filled parabolic band case for $A=0$, $U=0.5$ eV, $J=0.1$ eV, W (the bandwidth) $=1$ eV, $\alpha=0.2$ eV, and $\rho(E_F)=1.5$ states/eV. This curve is qualitatively similar to the experimental results for La_3S_4 and La_3Se_4,[12] and for TiH_{2-x}.[14] But it should be noted that the ratio given by Eq. (28) is found to be larger (~ 15) for La_3Se_4.[12] This might be due to the fact that close to $T=T_M$, the mean-field expression for e^2 in Eq. (26) as obtained from Eq. (17) is not correct, and the fluctuation effects have been found to be important. Also, a nonvanishing cubic term in e exists in the free energy (15) arising from the electrons other than the e_g electrons. Even if its value is small, this might cause the free energy to be nearly discontinuous very close to $T=T_M$. This then would make the derivative of χ below $T=T_M$ to be much sharper as has been observed for La_3S_4.[12]

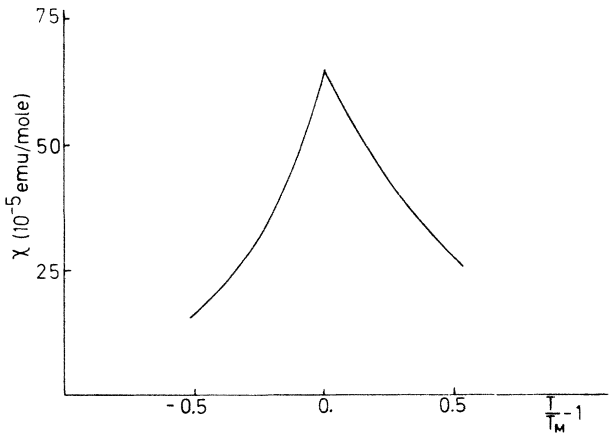

FIG. 2. Plot of magnetic susceptibility χ vs $(T/T_M)-1$ for the half-filled parabolic band.

V. ELASTIC CONSTANT

The variation of the elastic constant with temperature in degenerate band systems is of particular interest around $T=T_C$ and $T=T_M$. The expression for the total elastic constant C_s due to all the electrons is obtained from the second derivative of F with respect to e. In the cubic phase this gives

$$\frac{C_s(T)}{C_0}=\frac{1-(A+4\alpha)I_1}{1-AI_1}+2\alpha\left[\frac{B}{4}\right]^2\frac{I_1X_3m^2}{(I-AI_1)^2}\Theta(T_C-T) , \tag{29}$$

where

$$\Theta(T_C-T)=\begin{cases}1 & \text{for } T<T_C\\ 0 & \text{otherwise .}\end{cases}$$

So for $T>T_C$, C_s decreases with a decrease of temperature due to the first term in Eq. (29) and in the absence of any magnetic ordering would be zero in this model of a second-order phase transition at $T=T_M$, where $(A+4\alpha)I_1=1$. But if the magnetic interactions are sufficiently strong to cause magnetic ordering, then C_s would increase below T_C due to the second term in Eq. (29). The nature of variation of C_s on the two sides of T_C can be worked out by developing I_1 as a function of T/T_C-1, and we get the same ratio for the absolute value of the ratio of the derivatives of C_s on the two sides of T_C as given by Eq. (27). The increase below T_C is more rapid than the decrease above T_C. This variation for the half-filled-band case is shown in Fig. 3. The nature of the variation is again qualitatively similar to the recently published results on bcc iron.[4] It will be interesting to measure the temperature dependence of C_s in the weak ferromagnetic systems around $T=T_C$ so that the validity of the HF theory can be better judged.

We next consider the elastic constant around $T=T_M$. Since below T_M, e is nonvanishing, we get

$$\frac{C_s(T)}{C_0}=\frac{1-(A+4\alpha)I_1}{1-AI_1}+\frac{2\alpha I_1X_2(Ge)^2}{(1-AI_1)^4}\Theta(T_M-T) , \tag{30}$$

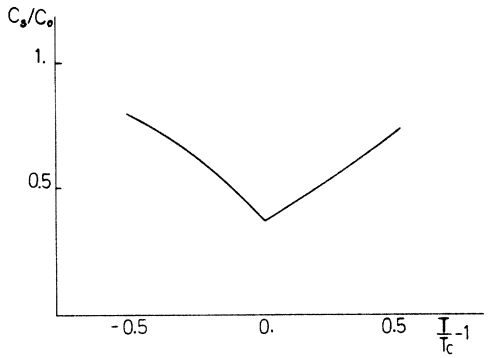

FIG. 3. Plot of C_s/C_0 vs $(T/T_C)-1$ for the half-filled parabolic band $A=0$, $U=0.8$ eV, $J=0.16$ eV, $W=1$ eV, $\alpha=0.15$ eV, and $\rho(\epsilon_F)=1.5$ states/eV.

where $\Theta(T_M-T)$ as already been defined. Therefore, in this Landau theory we obtain total softening of the elastic constant at $T=T_M$, and then C_s would increase due to the increasing values of e below T_M. The gradient of C_s on the two sides of T_M can similarly be calculated and is of the same form as that given by Eq. (27). Accurate measurements of C_s on La_3S_4 around $T=T_M$ would be useful to verify the present calculation. Available experimental results on the In-Tl alloy[18] for C_s on the two sides of T_M are in qualitative agreement with our analysis.

VI. MAGNETIC-FIELD DEPENDENCE OF T_M

The transition temperature T_M has been studied as a function of an external magnetic field H in La_3S_4 and La_3Se_4 and large values of $\Delta T_M/T_M^0$ [$\Delta T_M=T_M(H)-T_M^0$, T_M^0 being the value of T_M for $H=0$] have been obtained. Magnetic interactions can enhance this field dependence, and so it would be interesting to derive an expression of the enhancement factor from our free-energy expression (15). Since the elastic constant goes to zero at $T=T_M$, we derive C_s in presence and absence of H and finally get the following expression of ΔT_M:

$$\frac{\Delta T_M}{T_M^0}=-\frac{3}{2\pi^2}\frac{X_3}{X_1}\left[\frac{\mu_B H}{k_B T_M^0[1-B\rho(\epsilon_F)]}\right]^2 . \quad (31)$$

This shows clearly that a Stoner-type enhancement factor $[1-B\rho(\epsilon_F)]^{-2}$ is responsible for this enhancement of ΔT_M. Even if $B\rho(\epsilon_F)$ is not sufficient to cause magnetic ordering, this would decrease the value of T_M. For La_3S_4 the analysis of Pauli susceptibility and specific heat gives $B\rho(\epsilon_F)\simeq 0.58$, and consequently ΔT_M is enhanced by a factor of 6 and can well explain the observed value.

VII. SPECIFIC-HEAT JUMP AT $T=T_M$

From Landau's theory of phase transition one expects a jump in the value of the specific heat at $T=T_M$ and experimental results on La_3S_4 (Ref. 12) clearly indicate this. We shall derive an expression for the specific heat at $T=T_M$. The specific heat C_v is given by

$$C_v(T_M)=-T\frac{\partial^2 F}{\partial T^2}\bigg|_{T=T_M} . \quad (32)$$

From the minimization of F we get in the tetragonal phase

$$F=F_{PC}=-\frac{1}{4}\frac{a_2^2}{a_4} . \quad (33)$$

We make an expression of a_2 around $T=T_M$ and considering a_4 to be temperature independent but taking its value at $T=T_M$ we finally get the following expression for the change of specific heat at $T=T_M$:

$$\Delta C_v(T_M)=\tfrac{4}{3}\pi^4(A+4\alpha)\rho(\epsilon_F)^2(K_B T_M)^3 K_B\frac{X_1^2}{X_2} . \quad (34)$$

The effect of the magnetic interactions on ΔC_v only appear through A, and consequently it depends essentially on the elastic interactions involved in 4α and T_M. Also it depends strongly on $\rho(E_F)$.

VIII. DISCUSSIONS

We have studied the interplay between the elastic and magnetic interactions in a narrow twofold-degenerate band. Although for single-peaked DOS, the Landau expansion did not give possible coexistence of ferromagnetic and tetragonal phases, many of the physical properties of such a system are influenced by the two types of interactions. The analysis presented here on the basis of the HF theory gives results in qualitative agreement with the available experimental results but to get quantitative agreement it might be necessary to incorporate the local fluctuations of strains in addition to the spins which have been considered so far.

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