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Effect of pressure on the Curie temperature of Fe_3Pt

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We have performed self-consistent spin-polarized calculations for Fe_3Pt (which crystallizes in the Cu_3Au structure) at various Wigner-Seitz radii using the linear muffin-tin orbital atomic sphere approximation method. The average magnetic moment obtained at equilibrium lattice constant ($1.99\mu_B$) is close to the experimental value ($2.15\mu_B$). We have calculated the Curie temperature of this compound at various Wigner-Seitz radii, using the model proposed by Mohn and Wohlfarth. The quantities which are obtainable from the band-structure calculations are the only requisite for this model. The variation of the Curie temperature with Wigner-Seitz radii, i.e., with pressure, $T_C(P)$ is then compared with the experimental data.

I. INTRODUCTION

The magnetic properties of transition-metal intermetallic compounds have been the subject of great technological importance due to the numerous applications of permanent magnets in industry. From the theoretical point of view, the emphasis has been on elucidating the basic mechanism of magnetism. The understanding of magnetic properties is based on two approaches: mean-field theory (MFT) (Stoner model),[1] which stresses the itineracy and the spin-fluctuation-dependent description.[2] Spin-polarized self-consistent band-structure calculations (which are within MFT) have been very successful in calculating and predicting the magnetic moments of the magnetic intermetallic compounds. The calculated moments are in good agreement with experiments.[3] Although the calculation of the magnetic moment has received a lot of attention, the calculation of the Curie temperature T_C seems not to have attracted much attention. The theories of the T_C are given in the papers of Gunnarson[4] and Mohn and Wohlfarth (MW).[2] The main feature of MFT for T_C is the itinerant description of the electrons, originally assumed to be in a parabolic band, and the introduction of the exchange energy, which is proportional to the magnetization squared, the constant of proportionality being called the Stoner parameter. It has been derived in the Hartree-Fock approximation by Wohlfarth.[5] Gunnarson[4] used the Bloch representation for all the states, and to calculate the Stoner parameter, he applied the local spin density (LSD) approximation. This approach neglects the effect of localized, fluctuating moments which may be present in the system. The treatment of spin fluctuations in itinerant systems is described in MW's model in which they defined a characteristic temperature T_{SF} describing the influence of spin fluctuations. The theory then dwells on the relation between Stoner Curie temperature T_C^S, T_{SF}, and T_C. Both these theories require, as input, quantities which come out of a band-structure calculation.

Recently, there have been renewed attempts to calculate T_C of the rare-earth metals and compounds.[6–8] Jaswal[6] calculated the spin-polarized electronic structure for the rare-earth compounds $R_2Fe_{17}N_\xi$ (R=rare-earth metal) and demonstrated its relationship to site-dependent magnetic moments using the model by MW. Their calculation showed an increase in T_C for the nitrogenated materials in agreement with experiments. Brooks *et al.*[7] have done calculations for the T_C of rare-earth metals. They demonstrated the importance of including the exchange splitting of the $5d$ states in the treatment of magnetic properties under MFT and obtained comparatively better values for T_C, but still off from experimental values. Using this theory, Severin *et al.*[8] have calculated T_C for $GdCo_2$ and its hydride $GdCo_2H_4$. By simply scaling the calculated T_C of these two materials with the deGennes factor, they estimated the T_C for other heavy rare-earth cobalt compounds and their hydrides and obtained good agreement with experiment.

There have been fewer attempts to calculate T_C for transition metals and their compounds. Gunnarson[4] reported values of T_C, from the Stoner model within the LSD approximation, for Fe, Co, and Ni which were unrealistically high. Oles and Stollhoff[9] and Stollhoff[10] improved those results by treating the missing correlation effects using the local density approximation (LDA). Their results were still higher than experimental T_C values. However, MW's model of spin fluctuations could give value of T_C for Fe, Co, and Ni, close to experimental values. Also the calculated T_C for some transition-metal compounds (Y-Fe and Y-Co compounds[2]) were in agreement with experiment.

The band structure of Fe_3Pt has been calculated by numerous workers[11–13] and the agreement of ground-state properties (magnetic moment, density of states, electronic specific-heat coefficient) with experiment[14,15] is found to be good. Podgorny[11] has performed linear muffin-tin orbital (LMTO) calculations on Fe_3Pt under pressure and has shown that the magnetic moment variation can be correlated to the pressure dependence of the magnetic moment of Fe. Although experimental data for T_C and its pressure dependence is available,[16] no theoretical attempt has been made to calculate the same. Hence it would be interesting to perform such calculations especially since experimental data exist. In this paper we report calculations of T_C with MW's model. In Sec. II, method of calculation is given and results are discussed in Sec. III. In Sec. IV we summarize the conclusions.

II. METHOD OF CALCULATION

We have performed the band-structure calculations for the ordered compound Fe_3Pt using the linear muffin-tin orbital

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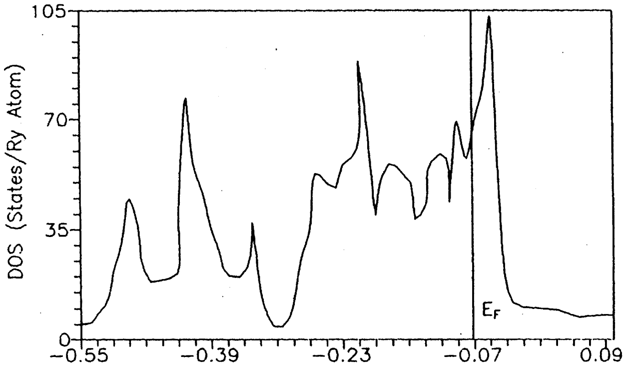

FIG. 1. Total density of states in paramagnetic Fe_3Pt. Fermi energy is marked (E_F).

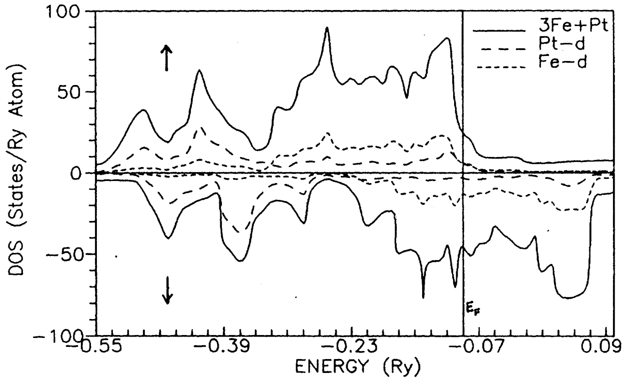

FIG. 2. d density of states for Pt,Fe atoms and total density of states in ferromagnetic Fe_3Pt.

method in the atomic sphere approximation (ASA) including the combined corrections to the ASA.[17] The parametrization of von Barth and Hedin[18] has been used for the exchange and correlation part of the effective one-electron potential, obtained within the LSD approximation to density functional theory. To include the important relativistic effects, the Dirac equation without the spin-orbit term has been solved to get the wave functions within atomic spheres. We have performed self-consistent calculations for a sample of 84 $\vec{k}$ points within 1/48th part of the irreducible Brillouin zone. The density of states (DOS) has been calculated using the tetrahedron method.[19]

To calculate T_C, we have used the model proposed by MW,[2] where they have derived an expression for calculating T_C,

$$\frac{T_C^2}{T_C^{S^2}} + \frac{T_C}{T_{\mathrm{SF}}} - 1 = 0, \tag{1}$$

where T_C^S, Stoner Curie temperature, is the temperature at which

$$I(E_F)\int_{-\infty}^{\infty} \frac{\partial f(E)}{\partial E} N(E)\,dE = 1, \tag{2}$$

where $N(E)$ is the DOS per atom per spin[4] and $I(E_F)$ is the Stoner parameter at the Fermi energy (E_F). T_{SF} is the characteristic temperature describing the influence of spin fluctuations and is given by

$$T_{\mathrm{SF}} = \frac{M_0^2}{10 k_B \chi_0}, \tag{3}$$

where M_0 and χ_0 are the magnetic moment and magnetic susceptibility of the system given as

$$M_0^2 = \frac{1}{N}\sum_{i=1}^{N} M_i^2 \quad \text{and} \quad \chi_0 = \frac{1}{N}\sum_{i=1}^{N} \chi_i . \tag{4}$$

N is the number of atoms in the unit cell. χ_i is calculated using the relation given by Wohlfarth[20] and Gersdorf,[21]

$$\chi_i^{-1} = \frac{1}{2\mu_B^2}\left[\frac{1}{2N^{\uparrow}(E_F)} + \frac{1}{2N^{\downarrow}(E_F)} - I\right]. \tag{5}$$

I is the Stoner parameter and $N^{\uparrow}(E_F)$ and $N^{\downarrow}(E_F)$ are spin-up and spin-down DOS per Fe atom. The Stoner theory, which takes into account only single-particle excitations, is valid for systems where T_{SF} is large compared with T_C. In a pure spin-fluctuation-dependent description of magnetic properties, the Stoner Curie temperature is always assumed to be infinite[22] and T_C is then equal to T_{SF}.

TABLE I. l-projected density of states at the Fermi energy, in states per Ry per unit cell at equilibrium WS radius for Fe_3Pt.

	PM		FM	
	Fe	Pt	Fe	Pt
s-DOS	0.072	0.520	0.137	0.144
p-DOS ↓	0.329	0.272	0.268	0.322
d-DOS	19.258	4.121	4.668	5.170
s-DOS			0.097	0.021
p-DOS ↑			0.646	0.887
d-DOS			13.572	3.604

III. RESULTS AND DISCUSSIONS

The experimental value of lattice constant, 3.73 Å,[23] corresponds to the equilibrium Wigner-Seitz (WS) radius equal to 2.7552 a.u. We have performed calculations at this value for the Cu_3Au structure compound Fe_3Pt. Calculations are done for paramagnetic (PM) and ferromagnetic (FM) phases. Figure 1 displays the total density of states for PM Fe_3Pt. Angular momentum projected DOS are given in Table I. The DOS at E_F [$N(E_F)$] is high, which actually causes the Stoner product $IN(E_F)$ to be greater than 1. Here I is the Stoner parameter for the compound, which is written as

$$I = \sum_{qll'} \left(\frac{N_{ql}}{N} I_{qll'} \frac{N_{ql'}}{N}\right), \tag{6}$$

where N and N_{ql} are the total DOS and site q and angular momentum l projected DOS at E_F, and $I_{qll'}$ are the intra-atomic exchange integrals. These are given by

TABLE II. WS radius, in a.u., and the magnetic moment for Fe_3Pt from various references, in μ_B per atom.

	From our calculations	From Ref. 11	From Ref. 12	Experiment[a]
WS radius	2.7552	2.77	2.77	2.75
Magnetic moment (Fe)	2.56	2.51	2.50	2.70
(Pt)	0.26	0.26	0.50	0.50
Average	1.99	1.95	2.00	2.15

[a] Reference 14.

$$I_{qll'} = \int_0^{S_q} r^2 K(r) \phi_l(E_F,r) \phi_{l'}(E_F,r) dr.$$

$K(r)$ is a function of the charge density,[24] $\phi_l(E_F,r)$ is the wave function of angular momentum l for states at E_F, and S_q is the atomic sphere radius.

Throughout our calculations we have used a frozen-core approximation in which the total energy contains the contribution from the valence states only. This approximation is valid at ambient and low pressures where the core states are highly localized. Since we are interested in the ground-state properties at ambient pressures, this approximation has been used. The total energy E of Fe_3Pt for PM and FM phases at the experimental equilibrium WS radius of 2.7552 a.u. is $E(\mathrm{PM}) = -188.254$ Ry and $E(\mathrm{FM}) = -188.295$ Ry, giving $E(\mathrm{FM}) < E(\mathrm{PM})$, which again confirms that Fe_3Pt is FM in nature. From Table I, we notice that at E_F most of the contribution is from Fe d states. Figure 2 displays the DOS curves for spin-up and spin-down Fe d and Pt d states and also the total DOS for FM Fe_3Pt is shown. The calculated average magnetic moment of $1.99\mu_B$ is found to be in close agreement with the experimental value of $2.15\mu_B$; however, moments on each site are slightly different (Table II). Podgorny[11] has obtained an average moment of $1.95\mu_B$ using the LMTO method and Hasegawa's value[13] is $2.0\mu_B$ using the APW method. Both of these calculations are at the theoretical equilibrium lattice constant, corresponding to a WS radius of 2.77 a.u., which is slightly different from that we have used. The differences in the magnetic moments may be attributed to the different convergence criteria and to the different number of $\vec{k}$ points used for Brillouin-zone integrations, besides the use of different WS radii. Table II shows the comparison of magnetic moments from different calculations.

The spin-fluctuation temperature T_{SF} is calculated using Eq. (3) and found to be 5979 K at the experimental equilibrium lattice constant. The value of Stoner Curie temperature T_C^S calculated by solving the integral (2) for different temperatures is 3438 K. According to the MW model T_C is thus given by Eq. (1). At the experimental lattice constant, we obtained T_C equal to 2588 K. The experimental value of T_C for Fe_3Pt is reported to be 400 K in Ref. 14 and 430 K in Refs. 25,26. Abd-Elmeguid and Micklitz[16] have experimentally investigated the temperature dependence of $\bar{B}_{\mathrm{eff}}$ (average magnetic hyperfine field) as a function of pressure, which in fact determines $T_C(P)$, i.e., T_C with pressure. Thus to check the effect of pressure on T_C we have performed self-consistent spin-polarized calculations for Fe_3Pt in the FM phase at different radii (2.4–2.7552 a.u.). Our magnetic moment variation with pressure shows a sharp monotonic decrease of magnetic moment with increasing pressure (Fig. 3), in agreement with the LMTO calculations of Podgorny.[11] Figure 4 shows the variation of $T_C(P)/T_C(0)$ with pressure, where $T_C(0)$ is the T_C at the experimental equilibrium lattice parameter. The experimental data[16] are also included in Fig. 4. An overall agreement between theory and experiment is noticed. Our calculations show a much more rapid decrease in T_C as a function of pressure than the experimental data.

The variation of T_C with pressure can arise from various factors: (i) variation of I with pressure, (ii) variation of the

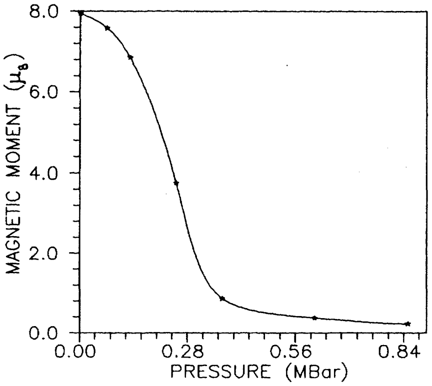

FIG. 3. Variation of magnetic moment with pressure.

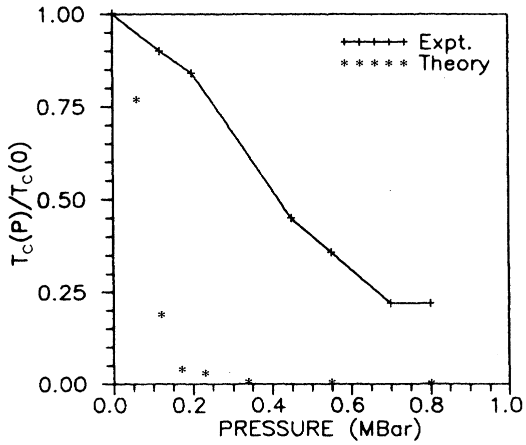

FIG. 4. Variation of $T_C(P)/T_C(0)$ with pressure (+ + + for experimental data and *** for theory).

DOS with pressure, or (iii) variation of the magnetic moment with pressure. Krasko[27] has shown that the variation of I with WS radius, i.e., with pressure, is needed to explain the metamagnetic behavior of Fe. For a variation of the WS radius from 2.75 to 2.5 a.u., an increase of 6% in I is reported. For the sake of completeness we have performed calculations by including the variation of I with pressure as suggested by Krasko and do not find any significant change in the calculated value of $T_C(P)/T_C(0)$. We have calculated I using Eq. (6) and found it to be constant (0.0176 Ry) for radii between 2.7552 and 2.675 a.u. The paramagnetic DOS decreases from 80.26 to 33.12 states/Ry per unit cell and is important in calculating T_C^S. However, the ferromagnetic DOS varies from 68.31 to 105.06 and then to 59.58 states/Ry per unit cell for radii 2.7552–2.6750 and 2.4000 a.u., respectively. Thus it is unlikely that the DOS can explain the variation of T_{SF} with pressure in Fe_3Pt. Our results seem to indicate that the rapid variation of T_C with pressure can be accounted for by the rapid variation of magnetic moment with pressure. This seems to corroborate the conclusion of Jaswal[6] and Severin *et al.*[8] In these calculations the magnetic moments do not change significantly; hence, good agreement was obtained with experiment. In our calculations the magnetic moment changes by more than factor of 40. This would seem to provide a more stringent test for MW theory. One important drawback of MW theory is that it does not address itself to the presence of high- and low-spin states. It would be essential to include this while discussing Fe compounds.

IV. CONCLUSIONS

We have calculated the pressure dependence of T_C for Fe_3Pt using the MW model with parameters taken from self-consistent band structure. Our calculations are in fair agreement with the experiment; however, exact agreement is lacking. We attribute the difference to the inadequacy of MW theory, which does not take into account the presence of high- and low-spin states in Fe. We believe that MW theory needs to be modified for better agreement between theory and experiment.

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