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Using a first-principles disordered-local-moment picture of itinerant-electron magnetism, we calculated the temperature and volume dependence of the magnetic moment and spin-spin correlations for fcc Fe in the paramagnetic state.

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Although the equilibrium crystal structure of ferromagnetic iron is body-centered-cubic (bcc), the magnetic properties of face-centered-cubic (fcc) iron are of abiding interest.[1] Most commonly, this arises from efforts to account for the stability of γ-iron[2] and the rich variety of magnetic structure displayed by Fe films grown on Cu substrates[3] or to understand the Invar phenomena in Ni_cFe_{1-c} and other similar alloys.[4] While we are mindful of such concerns, the work reported here has a different focus. We study fcc Fe because it provides us with a dramatic circumstance in which to address two of the central issues of itinerant-electron magnetism: the formation of magnetic moments and the nature of their interactions. We will present new evidence that as the lattice parameter, a, is reduced from 7.0 to 6.6 a.u. there is a transition from a high- to a low-moment state with the interaction between the moments changing from ferromagnetic (FM) to antiferromagnetic (AFM), respectively.

We shall limit our discussion to theoretical considerations alone. Recently, using the local-spin-density approximation (LSDA), a number of authors have studied the magnetic moment per unit cell as a function of the lattice spacing in both the FM and the AFM states.[5-7] The latest and most careful of these calculations is that of Wang, Klein, and Krakauer.[7] Their results are summarized in Fig. 1. For easy reference we also show the experimental lattice constant as a function of temperature. In agreement with previous calculations, they find in both phases a transition from a high- to a low-moment state as the lattice parameter is reduced. Moreover, their total-energy calculations imply that in the low-moment state antiferromagnetic order is preferred, while for the large-lattice-parameter, large-moment state, the energetically more favorable magnetic structure is ferromagnetic. Elsewhere Kübler[8] came to the same conclusions and made the interesting conjecture that one can expect a local moment, which is independent of its orientation with respect to its neighbors, only for those values of lattice constant a for which there are both FM and AFM solutions to the Kohn-Sham equations. In this Letter we study both the magnetic moments and the magnetic correlations as functions of lattice spacings in the paramagnetic disordered-local-moment (DLM) state. We also find that the moment collapses as the lattice spacing is reduced and thereby confirm the conjecture that the above phenomenon is independent of the overall magnetic structure. Moreover, by calculating the static **q**-dependent susceptibility we are able to conclude that at the same time as the moment col-

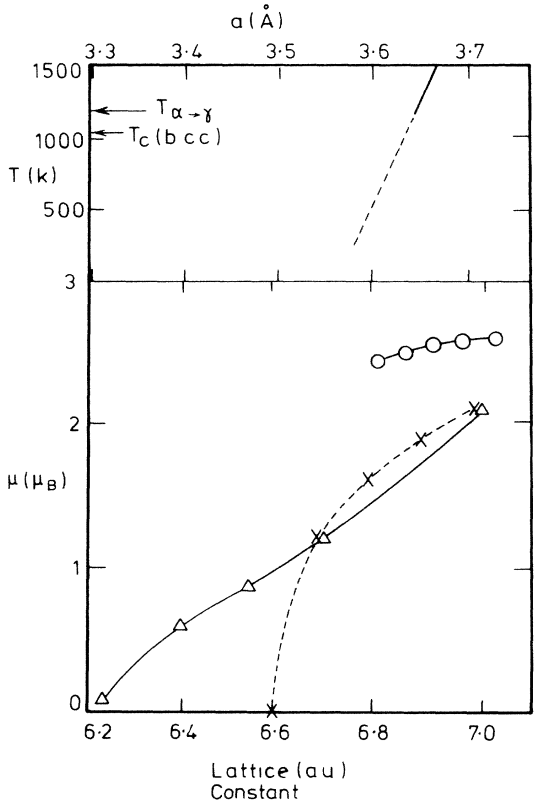

FIG. 1. The magnetic moments per site, μ, as calculated for the ferromagnetic (FM), antiferromagnetic (AFM), and disordered-local-moment (DLM) states of fcc Fe, denoted respectively by circles, triangles, and crosses. The results for the FM and AFM states are those by Wang, Klein, and Krakauer (Ref. 7) and those for the DLM state are of the present paper. The lines are added as guides to the eye.

lapses the interaction between moments changes from ferromagnetic to antiferromagnetic.

The DLM picture of Cyrot,[9] Hubbard,[10] and Hasegawa[11] has been reformulated in the language of spin-density-functional (SDF) theory by Gyorffy *et al.*[12] In this theory the electrons are treated within the LSD approximation but, instead of the usual FM or AFM case where all the unit cells are forced to be the same, an ensemble of states, whose spin polarization varies from site to site, is studied. The orientation of the spin polarization in the ith unit cell is depicted by the unit vector $\hat{\mathbf{e}}_i$ and an orientational configuration is described by the set $\{\hat{\mathbf{e}}_i\}$. The grand potential for the electron system whose spin polarization, averaged over each unit cell separately, is constrained to point along a set of prescribed local directions, $\{\hat{\mathbf{e}}_i\}$, is denoted by $\Omega(\{\hat{\mathbf{e}}_i\})$ and the total electronic free energy is written as $F=-\beta^{-1}\ln Z$, where

$$Z=\prod_i \int d^2\hat{\mathbf{e}}_i \exp[-\beta\Omega(\{\hat{\mathbf{e}}_i\})]. \tag{1}$$

In principle, $\Omega(\{\hat{\mathbf{e}}_i\})$ is to be calculated for each configuration by use of the LSD approximation. Fortunately, in the mean-field approximation for the statistical sum in Eq. (1), it is sufficient to evaluate the average of $\Omega(\{\hat{\mathbf{e}}_i\})$ with respect to the inhomogeneous product distribution function $P(\{\hat{\mathbf{e}}_i\})=\prod_i P_i(\hat{\mathbf{e}}_i)$. The situation is particularly tractable in the paramagnetic state where, without loss of generality, $\hat{\mathbf{e}}_i$ may be taken to be an Ising-type variable pointing only up or down, $\hat{e}_i^z=\pm 1$. Then $P_i(\hat{e}_i^z)$ may be parametrized by the average $m_i=\langle \hat{e}_i^z\rangle$ as follows: $P_i(\hat{e}_i^z)=\frac{1}{2}(1+m_i\hat{e}^z)$. Evidently, $\langle\Omega(\{\hat{e}_i^z\})\rangle$ is now a function of m_i. Two central results of the theory are that the Weiss molecular field h_i^{MF} and the spin-spin direct correlation function $S_{ij}^{(2)}\equiv\partial h_j^{\mathrm{MF}}/\partial m_i$ are given by

$$h_i^{\mathrm{MF}}=\left(\frac{\partial\langle\Omega\rangle}{\partial m_i}\right)_{\{0\}}, \quad S_{ij}^{(2)}=\left(\frac{\partial^2\langle\Omega\rangle}{\partial m_i\partial m_j}\right)_{\{0\}}, \tag{2}$$

respectively. Moreover, in the limit of rigid moments the spin-only susceptibility $\chi(\mathbf{q})$, in the paramagnetic state, is given by

$$\chi(\mathbf{q})=\tfrac{1}{3}\beta\bar{\mu}^2/[1-1/3\beta S^{(2)}(\mathbf{q})], \tag{3}$$

where $\bar{\mu}$ is the configurationally averaged magnetic moment and $S^{(2)}(\mathbf{q})$ is the lattice Fourier transform of $S_{ij}^{(2)}$. For a spin Hamiltonian in the random-phase approximation $\chi(\mathbf{q})$ is also given by Eq. (3) but with $S^{(2)}(\mathbf{q})$ replaced by the lattice Fourier transform of the exchange integral $J(\mathbf{q})$. Indeed, $S^{(2)}(\mathbf{q})$, although defined in fully itinerant terms by Eq. (2), may be interpreted as an effective interaction energy between the moments at i and j.

The above account of the magnetization fluctuations is only the first half of the theory. The second half deals with the calculation of h_i^{MF} and $S_{ij}^{(2)}$ on the basis of the SDF theory. Since the local orientations, $\hat{e}_i^z$, are to be regarded as independent random variables, the average $\langle\Omega\rangle$ may be calculated by use of the well known mean-field theory of disorder, namely the coherent-potential approximation (CPA). The relevant muffin-tin, charge self-consistent version of this Korringa-Kohn-Rostoker or KKR-CPA scheme was first used in the present context by Pindor *et al.*[13] and Oguchi, Terakura, and Hamada.[14] Yet again, in the spirit of the mean-field theory, one begins with the construction of the partially averaged, spin-dependent, local potential $\bar{v}_\sigma(\mathbf{r}-\mathbf{R}_i;\hat{e}_i^z)$ which depends only on the local random variable $\hat{e}_i^z$. It is calculated with the von Barth–Hedin version[15] of the LSD approximation. Then the corresponding Kohn-Sham equations are solved for the partially averaged Green's functions $\langle G_{\sigma,\sigma}(\mathbf{r},\mathbf{r};\epsilon)\rangle_{i,\hat{e}^z}$ by means of the KKR-CPA. From $\langle G_{\sigma'\sigma}(\mathbf{r},\mathbf{r};\epsilon)\rangle_{i,\hat{e}^z}$ the partially averaged charge densities $\bar{\rho}_\sigma(\mathbf{r};\hat{e}_i^z)$ are determined. This procedure is repeated until convergence in $\bar{v}_\sigma$. The local moment is given by

$$\bar{\mu}=\mu_{\mathrm{B}}\int_{\Omega_0} d^3r[\bar{\rho}_\uparrow(\mathbf{r};\hat{e}_i^z)-\bar{\rho}_\downarrow(\mathbf{r};\hat{e}_i^z)],$$

where Ω_0 is the unit cell. To find $S^{(2)}(\mathbf{q})$ one must consider a weakly inhomogeneous version of the above CPA scheme. In fact, $S_{ij}^{(2)}$ turns out to be a fairly typical linear-response function satisfying a Bethe-Salpeter–type integral equation.[11] In the calculations we shall report that we have solved both the self-consistent KKR-CPA problem for the local moment $\bar{\mu}$ and the integral equation for the response function $S^{(2)}(\mathbf{q})$ including all the matrix elements. To render the computations tractable we have worked at complex energies making full use of the extensive and powerful analytic properties of multiple-scattering theory.[16]

In the above DLM picture the dominant temperature dependence is due to the factor $\beta=1/k_{\mathrm{B}}T$ in the denominator of Eq. (3). It is the consequence of the entropy of the orientational fluctuations. However, $\bar{\mu}$ also depends on temperature through various Fermi factors. This effect is due to the entropy of thermally excited electron-hole pairs and, at the temperatures where we worked, is relatively insignificant. Therefore, in calculating $\bar{\mu}$ we have treated all the Fermi factors as step functions. On the other hand, in the calculations of $S^{(2)}(\mathbf{q})$ all the electron-hole pair effects were treated properly by our summing over the appropriate Matsubara frequencies numerically.

Our results for the local moments are indicated by crosses in Fig. 1. Note that, in general, the size of $\bar{\mu}$ depends on the orientation of the environment. While in the FM state all nearest neighbors are parallel, in the type-I AFM state (CuAu structure), used in Ref. 7, there are eight parallel and four antiparallel nearest

neighbors. In the DLM state the most common nearest-neighbor environment is six up and six down. Thus, in general, $\bar{\mu}^{DLM}$ should be quite different from μ^{FM} but close to μ^{AFM}. As is clear from Fig. 1, this is in fact what we find. The remarkable agreement between our DLM results and the AFM moments of Wang *et al.*[7] should be taken as evidence that the DLM picture, when implemented within the framework of SDF theory, can yield quantitative results which are directly comparable to those for ordered orientational arrangements.

Having confirmed that the local moment on fcc Fe collapses as the lattice parameter shrinks from 7.0 to 6.6 a.u., we now go on to study the nature of the interactions between these moments. We do this by calculating the wave-vector–dependent static susceptibility $\chi(\mathbf{q})$ using the mean-field theory outlined above.

Anticipating that the small moments of fcc Fe, like those of Ni, will not be rigid, we have used a more general formulation of the problem than the arguments leading to Eq. (3). It allows the local moment to change in response to both the external magnetic field, directly, and the change in the environment due to the increased number of up moments produced by the external field. An outline of this theory has been given by Staunton *et al.*[17]

The results of our calculations of $\chi(\mathbf{q})$, along the [111] direction, for various lattice spacings and at $T=1400$ K, are shown in Fig. 2. For each value of the lattice parameter, we also show $\bar{\mu}^2/3k_BT$ which is both the high-temperature limit of $\chi(\mathbf{q}=0)$ and the susceptibility of the noninteracting moments. Evidently, any deviation of $\chi(\mathbf{q})$ from this constant value should be interpreted as being due to correlations between the moments. A maximum at $\mathbf{q}=0$ implies that the paramagnetic state is potentially unstable to spontaneous formation of a uniformally magnetized state. Namely, as the temperature is lowered this peak will grow to infinity at a temperature T_c where a phase transition to a ferromagnetic state occurs. A maximum of $\chi(\mathbf{q})$ elsewhere in the Brillouin zone implies incipient antiferromagnetic phases. These may be of different kinds. Type-I antiferromagnetism is associated with the Lifshitz special point[18] $\mathbf{q}_0=(2\pi/a)(1,0,0)$ and its star. It consists of alternating ferromagnetic (100) planes of opposite polarizations. On the other hand, type-II antiferromagnetism is signaled by an instability at $\mathbf{q}_0=\pi/a\,(1,1,1)$ corresponding to alternating [111] ferromagnetic planes of opposite polarization.

Clearly, the most striking feature of Fig. 2 is the shift of weight from $\mathbf{q}=0$ to finite $\mathbf{q}$ as the lattice parameter, a, is reduced. This we interpret as a reflection of the fact that the magnetic correlations are changing from ferromagnetic to antiferromagnetic. Indeed, at $a=6.7$ a.u. the maximum is at the zone

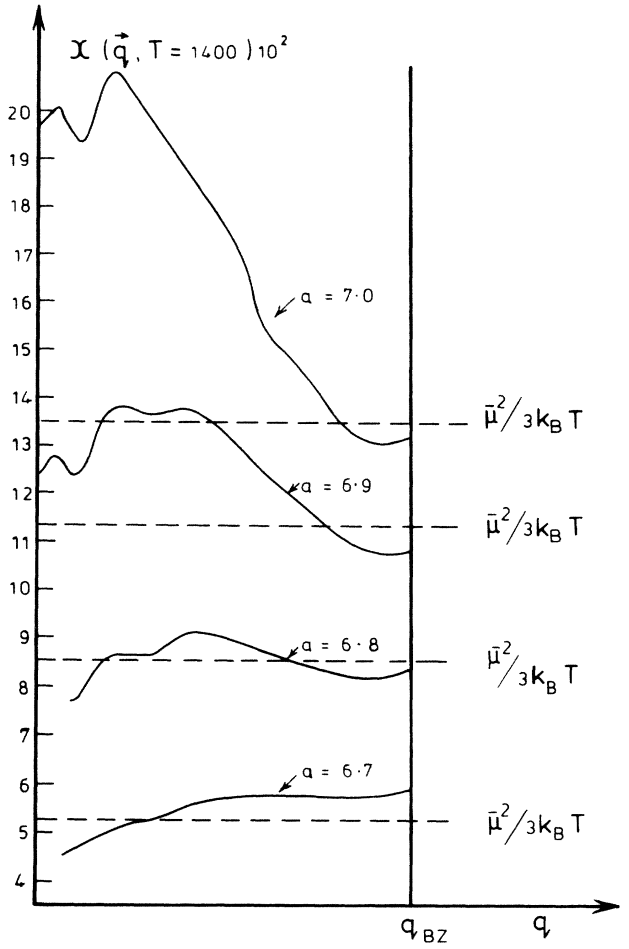

FIG. 2. The static susceptibility $\chi(\mathbf{q})$ at $T=1400$ in the [111] direction for fcc Fe at 7.0, 6.9, 6.8, and 6.7 a.u. For $q<q_{BZ}$ the computational difficulties are such that no definite conclusion is possible as to whether the center of the main peak is at $\mathbf{q}=0$ or not. For the sake of our discussion we have assumed it to be at $\mathbf{q}=0$.

boundary. Moreover, calculations for several directions were performed, which are not reported here, and the special point $(\pi/a)(1,1,1)$ is the absolute maximum. This suggests a low-temperature phase with type-II magnetic structure.

In Fig. 1 we indicate the range of lattice constants for which γ-iron exists at high temperatures. For these values of lattice parameters Brown *et al.*[19] studied $\chi(\mathbf{q})$ in neutron scattering experiments, using polarization analysis. We note with interest that they found the forward scattering peak in $\chi(\mathbf{q})$ much broader in fcc than in bcc Fe. Comparing the results reported here with those of Staunton *et al.*[17] for bcc Fe we observe the same effect.

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