Onsager Cavity Fields in Itinerant-Electron Paramagnets

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For an itinerant system of electrons we develop a first-principles theory of spin fluctuations. It is based on the idea of a generalized Onsager cavity field which varies slowly compared to the motion of individual electrons. It gives an account of both the good local moment and the weak itinerant ferromagnet limits within the same framework. We illustrate its consequences by explicit calculations for Fe and Ni.

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One of the oldest problems in magnetism is its nature in transition metals. While the modern version of the Stoner model [1], namely, spin-polarized band theory based on the local-spin-density approximation [2], gives a reasonable description of the magnetic ground state in most metals, above the Curie temperature T_C it fails to account for the local moments observed in many neutron-scattering experiments [3]. This obvious shortcoming of the theory was remedied, in principle, some ten years ago, by the introduction of local-moment-like thermal spin fluctuations into the itinerant model of electrons [4]. Unfortunately the *ab initio* implementation of this disordered local moment (DLM) picture [5] works well for Fe but not for Ni. In this Letter we go beyond the mean-field (MF) approximation used in working out the consequences of the disordered local moment picture by replacing the Weiss field in the calculation with a generalized Onsager cavity field [6]. As we shall show presently the new theory greatly improves the description of both Fe and Ni and is couched in fully electronic terms. In other words, it gives a fair account of the spin fluctuations in both the good-moment and the weak itinerant ferromagnetic [7] limits.

In the interest of clarity we begin with a brief outline of Onsager's arguments as they are appropriate for the classical Heisenberg model, $H=-\sum_{ij}J_{ij}\hat{\mathbf{e}}_i\cdot\hat{\mathbf{e}}_j$, where $\hat{\mathbf{e}}_i$ is a classical unit vector. Following the discussion of Brout and Thomas [6], we calculate the magnetization $\mathbf{m}_i=\langle\hat{\mathbf{e}}_i\rangle$ at each site i due to a small magnetic field $\mathbf{h}_j$ at each site j in the paramagnetic state. In the mean-field approximation, to linear order in $\mathbf{h}_j$,

$$\mathbf{m}_i=\frac{\beta}{3}\left[\sum_j J_{ij}\mathbf{m}_j+\mathbf{h}_i\right], \tag{1}$$

where $\beta=1/k_BT$. Found wanting, Onsager sought to improve on the MF approximation by observing that the Weiss field $\mathbf{h}_i^W$ on the right-hand side of Eq. (1) should not include the effects of the magnetization on the site i. Clearly this implies that we should replace Eq. (1) by

$$\mathbf{m}_i=\frac{\beta}{3}\left[\sum_j J_{ij}(\mathbf{m}_j-\delta\mathbf{m}_j^{(i)})+\mathbf{h}_i\right], \tag{2}$$

where the term in square brackets is now called the cavity field $\mathbf{h}_i^c$. To complete the theory one must now calculate the induced magnetization $\delta\mathbf{m}_j^{(i)}$ in terms of the set $\{\mathbf{m}_l\}$. A way to proceed is to note that for a small cavity field, $\delta\mathbf{m}_j^{(i)}=\tilde{\chi}_{ji}\mathbf{h}_i^c$, where $\tilde{\chi}_{ji}$ is the yet unknown full susceptibility tensor $\tilde{\chi}_{ji}=\delta\mathbf{m}_j/\delta\mathbf{h}_i$. Moreover, $\mathbf{m}_i=\tilde{\chi}_{ii}\mathbf{h}_i^c$ and hence the cavity field $\mathbf{h}_i^c$ may be written as $\mathbf{h}_i^c=\tilde{\chi}_{ii}^{-1}\mathbf{m}_i$. Consequently, $\delta\mathbf{m}_j^{(i)}=\tilde{\chi}_{ji}\tilde{\chi}_{ii}^{-1}\mathbf{m}_i$. Substituting this result into Eq. (2) and taking the derivative of both sides with respect to $\mathbf{h}_j$ yields a closed equation for the susceptibility tensor $\tilde{\chi}_{ij}$ whose matrix elements are $\chi_{ij}^{\alpha\beta}$, where $\alpha,\beta=x,y,z$. In terms of the lattice Fourier transforms $J(\mathbf{q})$ and $\tilde{\chi}(\mathbf{q})$ of J_{ij} and $\tilde{\chi}_{ij}$, respectively, this reads

$$\tilde{\chi}(\mathbf{q})=(\beta/3)\{[J(\mathbf{q})\tilde{1}-\tilde{\lambda}]\cdot\tilde{\chi}(\mathbf{q})+\tilde{1}\}, \tag{3}$$

$$\tilde{\lambda}=\tilde{\chi}_{ii}^{-1}\int d\mathbf{q}\,J(\mathbf{q})\tilde{\chi}(\mathbf{q}). \tag{4}$$

For the Hamiltonian at hand the above result is sometimes referred to as the mean spherical approximation and is well known to represent an improvement on the MF approximation. In what follows we present an analogous generalization of the first-principles mean-field DLM theory of Oguchi, Terakura, and Hamada, Pindor *et al.* [5], Gyorffy *et al.* [8], and Staunton *et al.* [9].

We start with the assumption that, on a time scale τ long compared with the relevant inverse bandwidth, the spins of the individual electrons are sufficiently correlated to leave the magnetization averaged over a time τ, and the unit cell, nonzero. The orientations of these local magnetizations $\{\hat{\mathbf{e}}_i\}$ vary slowly in time while their magnitude fluctuates rapidly on the time scale of τ. Their average over τ is defined as the local moment and it changes with the orientational configuration, i.e., $\mu_k=\mu_k\{\hat{\mathbf{e}}_i\}$. The standard spin-density-functional theory for studying electrons in spin-polarized metals can be adapted to describe the states of the system for each orientational configuration $\{\hat{\mathbf{e}}_i\}$. In principle such a description yields local moments $\mu_k\{\hat{\mathbf{e}}_i\}$ and the electronic grand potential $\Omega\{\hat{\mathbf{e}}_i\}$ for the constrained system. Thus the long-time averages can be replaced by ensemble averages with the Gibbsian measure $P\{\hat{\mathbf{e}}_i\}=Z^{-1}\exp(-\beta$

$\times\Omega\{\hat{\mathbf{e}}_i\})$, where the partition function $Z=\prod_i\int d\hat{\mathbf{e}}_i \times\exp(-\beta\Omega\{\hat{\mathbf{e}}_i\})$ and the thermodynamic free energy, which accounts for the entropy associated with the orientational fluctuations as well as creation of electron-hole pairs, is given by $F=-k_BT\ln Z$.

In the previous implementations of the DLM picture [5,8,9] the above scheme was rendered tractable by evaluating Z in the MF approximation and using the self-consistent-field Korringa-Kohn-Rostoker coherent-potential-approximation (SCF-KKR-CPA) method [10] to approximate the Weiss field. Explicit calculations for bcc Fe yielded $\langle\mu_i\{\hat{\mathbf{e}}_j\}\rangle_{\hat{\mathbf{e}}_i}=\mu_i(\hat{\mathbf{e}}_i)=\bar{\mu}=1.91\mu_B$, where the partial average $\langle\mu_i\{\hat{\mathbf{e}}_j\}\rangle_{\hat{\mathbf{e}}_i}$ means the average over all configurations with the specific orientation $\hat{\mathbf{e}}_i$ at the site i. While this result is in good quantitative agreement with experiment, for Ni $\bar{\mu}$ was found to be zero and the theory reduced to the conventional Stoner model with all its shortcomings.

We shall now improve the above theory by treating the local magnetization as a response to the Onsager cavity field rather than the Weiss field of the MF approximation. Once again a small magnetic field $\{\mathbf{h}_i\}$ is applied to the paramagnetic system. This induces a small deviation $\delta P_i(\hat{\mathbf{e}}_i)$ from the equilibrium single-site distribution function $P_i^0(\hat{\mathbf{e}}_i)=1/4\pi$. As a consequence the local magnetization $\mathbf{M}_i=\int d\hat{\mathbf{e}}_i\,\mu_i(\hat{\mathbf{e}}_i)\hat{\mathbf{e}}_iP(\hat{\mathbf{e}}_i)$ may be written as a sum of two parts, $\mathbf{M}_i=\boldsymbol{\mu}_i+\bar{\mu}\mathbf{m}_i$,

$$\boldsymbol{\mu}_i=\frac{1}{4\pi}\int d\hat{\mathbf{e}}_i\,\delta\mu_i(\hat{\mathbf{e}}_i)\hat{\mathbf{e}}_i\,, \tag{5}$$

$$\mathbf{m}_i=\int d\hat{\mathbf{e}}_i\,\hat{\mathbf{e}}_i\delta P_i(\hat{\mathbf{e}}_i)\,, \tag{6}$$

and correspondingly the susceptibility takes the form $\tilde{\chi}_{ij}=\tilde{\chi}_{ij}^{\mu}+\tilde{\chi}_{ij}^{m}$. Evidently the first term $\boldsymbol{\mu}_i$ describes how the magnitude of the local moments responds to the external field, whereas the second term $\mathbf{m}_i=\langle\hat{\mathbf{e}}_i\rangle$ describes how they tend to align with the field.

If we apply the procedure in the introduction and calculate the response of the CPA medium to the Onsager cavity field and subtract those changes, $\{\delta\boldsymbol{\mu}_l^{(i)}\}$, $\{\bar{\mu}\delta\mathbf{m}_l^{(i)}\}$, that are derived from the induced magnetization at the site i, we find for a paramagnetic system,

$$\bar{\mu}\mathbf{m}_i=\frac{\beta}{3}\left(\sum_{l\neq i}[\bar{\mu}S_{il}^{\mathbf{mm}}(\mathbf{m}_l-\delta\mathbf{m}_l^{(i)})+\bar{\mu}S_{il}^{\mathbf{m}\mu}(\boldsymbol{\mu}_l-\delta\boldsymbol{\mu}_l^{(i)})+\bar{\mu}\Sigma_{il}\mathbf{h}_l]+\bar{\mu}^2\mathbf{h}_i\right), \tag{7}$$

where, as in Ref. [9],

$$S_{il}^{m_\alpha,m_\beta}=\delta^2\Omega^{\rm CPA}/\delta m_\alpha\delta m_\beta=S_{il}^{mm}\delta_{\alpha\beta}\,,\quad S_{il}^{m_\alpha,\mu_\beta}=\delta^2\Omega^{\rm CPA}/\delta m_\alpha\delta\mu_\beta=S_{il}^{m\mu}\delta_{\alpha\beta}\,,$$

and

$$\Sigma_{il}^{\alpha\beta}=\delta^2\Omega^{\rm CPA}/\delta m_\alpha\delta h_\beta=\Sigma_{il}\delta_{\alpha\beta}\,,$$

and $\Omega^{\rm CPA}$ is the grand potential for an inhomogeneous CPA medium averaged over all configurations. The SCF-KKR-CPA method also provides an expression for $\mu_i(\hat{\mathbf{e}}_i)$, the magnitude of the partially averaged moment on the site i oriented along $\hat{\mathbf{e}}_i$,

$$\mu_i(\hat{\mathbf{e}}_i)=-\mathrm{Im}\pi^{-1}\int d\varepsilon f(\varepsilon-\nu)\int d\hat{\mathbf{e}}_i\,\mathrm{tr}\sigma\cdot\hat{\mathbf{e}}_i\int d\mathbf{r}_i\langle\tilde{G}(\mathbf{r}_i,\mathbf{r}_i;\varepsilon)\rangle_{\hat{\mathbf{e}}_i}\,,$$

where f is the Fermi function, ν is the electronic chemical potential, $\sigma^x,\sigma^y,\sigma^z$ are the Pauli spin matrices, and $\langle\tilde{G}\rangle_{\hat{\mathbf{e}}_i}$ is the partially averaged Green function. Using this expression as a starting point it is easy to derive the following Onsager corrected equation:

$$\begin{aligned}\boldsymbol{\mu}_i&=\frac{1}{\pi}\int d\hat{\mathbf{e}}_i\,\delta\mu_i(\hat{\mathbf{e}}_i;\{\mathbf{m}_l-\delta\mathbf{m}_l^{(i)}\},\{\boldsymbol{\mu}_l-\delta\boldsymbol{\mu}_l^{(i)}\})\hat{\mathbf{e}}_i\\&=\sum_{l\neq i}\gamma_{il}^{\mu\mathbf{m}}(\mathbf{m}_l-\delta\mathbf{m}_l^{(i)})+\sum_l\gamma_{il}^{\mu\mu}(\boldsymbol{\mu}_l-\delta\boldsymbol{\mu}_l^{(i)})+\sum_l\chi_{il}^0\mathbf{h}_l\,,\end{aligned} \tag{8}$$

where $\gamma_{ij}^{\mu_\alpha m_\beta}=\delta\mu_i^\alpha/\delta m_j^\beta=\gamma_{ij}^{\mu m}\delta_{\alpha\beta}$, $\gamma_{ij}^{\mu_\alpha\mu_\beta}=\delta\mu_i^\alpha/\delta\mu_j^\beta=\gamma_{ij}^{\mu\mu}\times\delta_{\alpha\beta}$, and χ_{ij}^0 is the Pauli susceptibility.

Clearly Eqs. (7) and (8) are the analog of Eq. (2) in the present case where both the orientation and the size of the moments fluctuate. Moreover the direct correlation functions S^{mm}, $S^{m\mu}$, $\gamma^{\mu m}$, and $\gamma^{\mu\mu}$ as well as χ^0 and Σ are available from the SCF-KKR-CPA calculations performed in the paramagnetic state. Thus we may proceed with the theory as in the case of the simple Heisenberg model. To do so we suppose that $\bar{\mu}\delta m_l^{(i)}=\chi_{li}^m h_i^c=\chi_{li}^m\chi_{ii}^{-1}M_i$ and $\delta\mu_l^{(i)}=\chi_{li}^\mu h_i^c=\chi_{li}^\mu\chi_{ii}^{-1}M_i$, where M_i as before is the magnitude of the total magnetization on the site i and the tensors such as $\tilde{\chi}_{li}=\chi_{li}\tilde{1}$ are now expressed in terms of scalar quantities in the paramagnetic state. Using these assumptions and Eqs. (7) and (8) we arrive at the following analogs of Eq. (3):

$$\chi^m(\mathbf{q})=(\beta/3)\{S^{mm}(\mathbf{q})\chi^m(\mathbf{q})+\bar{\mu}S^{m\mu}(\mathbf{q})\chi^\mu(\mathbf{q})-\Lambda_1[\chi^m(\mathbf{q})+\chi^\mu(\mathbf{q})]+[\bar{\mu}^2+\bar{\mu}\Sigma(\mathbf{q})]\}\,, \tag{9}$$

$$\chi^\mu(\mathbf{q})=\gamma^{\mu m}(\mathbf{q})\chi^m(\mathbf{q})+\gamma^{\mu\mu}(\mathbf{q})\chi^\mu(\mathbf{q})-\Lambda_2[\chi^m(\mathbf{q})+\chi^\mu(\mathbf{q})]+\chi^0(\mathbf{q})\,, \tag{10}$$

with $\Lambda_1=\chi_{ii}^{-1}\int d\mathbf{q}[S^{mm}(\mathbf{q})\chi^m(\mathbf{q})+\bar{\mu}S^{m\mu}(\mathbf{q})\chi^\mu(\mathbf{q})]$ and $\Lambda_2=\chi_{ii}^{-1}\int d\mathbf{q}[\gamma^{\mu m}(\mathbf{q})\chi^m(\mathbf{q})+\gamma^{\mu\mu}(\mathbf{q})\chi^\mu(\mathbf{q})]$. On integrating

Eqs. (9) and (10) over $\mathbf{q}$ we find $\chi_{ii}=\beta\bar{\mu}^2/3+\chi_{ii}^0$ from which a high-temperature estimate of the magnetic correlations can be extracted and worked out to be $\langle M_i^2\rangle\approx\bar{\mu}^2+3\chi_{ii}^0/\beta$.

For the rigid local-moment system in which itinerancy effects are small $S^{m\mu}$, $\gamma^{\mu m}$, $\gamma^{\mu\mu}$, and χ^0 vanish, and we obtain the classical Heisenberg results with "spins" of length $\bar{\mu}$. As shown by Ref. [6], the approach is equivalent to the spherical model. On the other hand, in a system where no "local moment" is set up on the average in the paramagnetic phase, $\bar{\mu}=0$ and S^{mm}, $S^{m\mu}$, and $\gamma^{\mu m}$ also vanish and $\chi^m(\mathbf{q})=0$, $\chi(\mathbf{q})=\chi^\mu(\mathbf{q})=\chi^0(\mathbf{q})/[1-\gamma^{\mu\mu}(\mathbf{q})+\Lambda_2]$ with $\Lambda_2=(\chi_{ii}^0)^{-1}\int d\mathbf{q}\,\gamma^{\mu\mu}(\mathbf{q})\chi^\mu(\mathbf{q})$. Note that $\gamma^{\mu\mu}$ is a product of a Stoner exchange-correlation term and a Pauli susceptibility χ^0. The theory now has the form of the static, high-temperature limit of the theory of Moriya and co-workers, Lonzarich and Taillefer, and others [7] to describe weak itinerant ferromagnets. The interactions between spin fluctuations are dealt with self-consistently through Λ_2.

It is evident that the dynamical effects of the spin fluctuations have been omitted thus far. As emphasized by Moriya and co-workers and others [7], their treatment is necessary for a full description of the neutron-scattering data and the properties of a range of weak itinerant ferromagnets. We point out, however, that the theory described here, with its single assumption of a time-scale separation between the fast orbital motions and slower spin fluctuations, is set up consistently within the confines of spin-density-functional theory. It is uniquely parameter free, and being based on the generalization of the Onsager cavity field, it represents a well-defined stage of approximation. Once its scope of validity is established it will provide an appropriate basis for future *ab initio* developments.

Figure 1(a) shows a comparison between theory and experiment [11] of the static uniform susceptibility for bcc iron [the experimental values are for Fe(5.7%Si)]. The Curie temperatures T_C of 1015 and 1040 K are in very good agreement (the "mean-field" estimate was 1280 K [8,9]). For temperatures in excess of T_C+300 K, both fit Curie-Weiss (C-W) laws $\chi=C/(T-\theta)$, each with $\theta\approx1100$ K. The effective moments, μ_{eff} ($C=\mu_{\text{eff}}^2/3$), of $1.96\mu_B$ and $3.13\mu_B$, respectively, are not in such good agreement, although the theoretical value is slightly enhanced over the "local" moment value of $1.91\mu_B$. On analyzing the $\mathbf{q}$ dependence of the susceptibility we find that at $1.25T_C$, for $|\mathbf{q}|\le1$ Å^{-1}, $\chi(\mathbf{q})$ varies as $\chi(0)/(1+q^2/\kappa^2)$ with the inverse correlation length $\kappa\approx0.37$ Å^{-1}. (This is similar to analogous results for a nearest-neighbor Heisenberg model with the same T_C.) This value compares favorably with $\kappa\approx0.4$ Å^{-1} quoted by Shirane, Boni, and Wicksted [12] who interpreted their inelastic neutron-scattering experiments in terms of simple paramagnetic scattering. In summary, we find that a Heisenberg model provides a qualitative description of bcc iron.

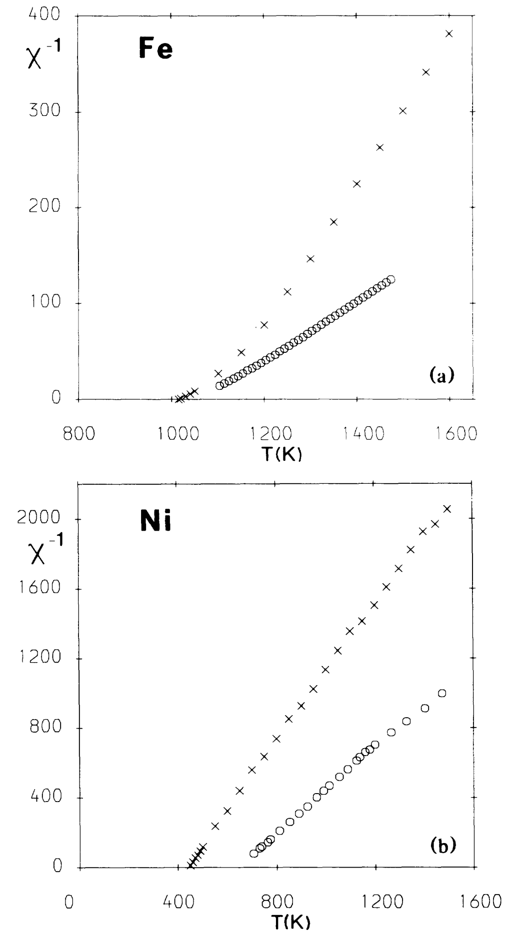

FIG. 1. (a) The temperature dependence of the inverse paramagnetic susceptibility of bcc iron in atomic units. The crosses show the theoretical results; circles show the experimental data [11]. (b) The temperature dependence of the inverse paramagnetic susceptibility of fcc nickel in atomic units. The crosses show the theoretical results; circles show the experimental data [11].

Recall that the earlier mean-field theory [5,8,9] could not give a satisfactory treatment for nickel. The new theory outlined here, however, is able to provide a reasonable account of experimental data. We find no "local" moment, $\bar{\mu}=0$, and $\chi(\mathbf{q})$ is determined solely by $\chi^\mu(\mathbf{q})=\chi_0(\mathbf{q})/[1-\gamma^{\mu\mu}(\mathbf{q})+\Lambda_2]$, $\Lambda_2=(\chi_{ii}^0)^{-1}\int d\mathbf{q}\,\gamma^{\mu\mu}(\mathbf{q})\times\chi^\mu(\mathbf{q})$. Figure 1(b) shows the theoretical-experimental [11] comparison. Both show approximate C-W behavior. Although at 450 K and some 200 K lower than the experimental value, T_C is substantially reduced from the Stoner value of 3000 K. Effective moments of $1.21\mu_B$ from theory and $1.6\mu_B$ from experiment are also in fair agreement. Evidently there is little connection with the low-temperature saturated magnetization per atom of $0.6\mu_B$ as accorded by a Rhodes-Wohlfarth plot [13]. For

$|\mathbf{q}| \leq 1$ Å^{-1} the wave-vector-dependent susceptibility $\chi(\mathbf{q}) \approx \chi(0)/(1+q^2/\kappa^2)$, where κ^2 has roughly the same temperature dependence as $\chi(0)^{-1}$, a feature which has also been noted from experiment [12]. At $T=1.25T_C$, $\kappa \approx 0.28$ Å^{-1} which agrees well with the inverse spin correlation length of ≈ 0.22 Å^{-1} extracted from the simple paramagnetic scattering analysis of Shirane, Boni, and Wicksted [12] of their inelastic neutron-scattering data.

In summary, we have presented a first-principles framework for Onsager cavity fields in itinerant-electron systems and at this point add that the approach is sufficiently general to deal with a variety of slowly varying fluctuations such as those appropriate to composition and strain in metal alloys. The first application on spin fluctuations in the paramagnetic states of iron and nickel finds that iron is related superficially to a Heisenberg model but nickel can be analyzed in terms of traditional Stoner theory although spin fluctuations have drastically renormalized the exchange interaction and lowered T_C.

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