Spectral statistics on itinerant magnetism in nanoscale metallic grains

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Abstract

Effect of the spectral statistics and level correlation on the itinerant ferromagnetism (FM) in nanoscale metallic grains is presented. The magnetization and spin susceptibility at zero temperature depend only on the spin splitting and the average level spacing. The Stoner criterion implies that the Coulomb interaction is greater than the Fermi energy for the appearance of FM long-range order. The spin wave excitation is found to be different from that of translation-invariant lattice. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

With the development of the nanotechnology and nanostructure devices, many distinguished properties are found in magnetic nanostructures which may provide potential applications in technique, for example, the spin properties of nanostructure is applied for single-electron transistor and quantum computation [1]. Study on these novel systems also has fundamental meaning for understanding the electronic correlation in nanoscopic and mesoscopic systems. The semiconductive and metallic nanostructures have attracted much theoretical and experimental attention in the past decade [2,3]. In contrast to the finite-size insulating Heisenberg ferromagnets (FM), the magnetic properties of metallic grains are very sensitive to the distribution of energy level and electronic states in these nanoscale grains. At sufficiently low-temperature $k_B T < \delta_{\text{av}}$, $\delta_{\text{av}}$ being the average level spacing of the metallic grain, the electronic states are dominated by the mesoscopic quantum effects; the many electron correlation characters are manifested by the quantum interference and the spectral statistics. However, the quantum interference of electrons becomes unimportant in many-spin grains due to the elimination of quantum interference, which is similar to the destruction of quantum interference effect under magnetic field. Therefore, the spectral statistics arising from the disorder or rough boundary dominate the magnetic properties of metallic grains. In this paper, we consider the clean irregular metallic grain, which corresponds to the ballistic case.

The goal of this paper is to elucidate how spectral statistics affect the low-temperature metallic magnetism and spin wave dynamics. The appearance of FM long-range order in the nanoscale grains is defined as one where the spin correlation is all over the system and the rotation symmetry of the spin is broken. Starting from the Hubbard model and in the self-consistent mean-field approximation, we find in the nanoscale metallic grains, the spin exchange splitting and the average level spacing determining the zero-temperature magnetic and spin wave properties, and the Stoner criterion for the appearance of FM long-range order associating with the Fermi energy.

2. Zero-temperature magnetic properties

We start from the Hubbard model in the presence of magnetic field $B$:

$$
H = \sum_{k, \sigma} (\epsilon_k - \sigma \mu_B B) c_{k\sigma}^\dagger c_{k\sigma} \\
+ \frac{U}{2N_{k+q, \sigma}} c_{k+q, \sigma}^\dagger c_{k, \sigma} c_{k-1, \sigma}^\dagger c_{k, \sigma},
$$

(1)
where $c_{L_n}$ creates an itinerant electron (IE), $U$ describes the on-site Coulomb interaction and $\varepsilon_k$ the spectrum of IE in the metallic grains. Since translation invariance is broken in nanoscale grains, the wave vector $k$ does not represent the quasi-momentum of IEs; here it is used as the index of the positions of the energy levels for convenience.

The spectrum of the IE of the metallic grain ensemble with disorder or boundary irregularity at low temperature will be distributed in terms of Wigner’s random matrix theory [4–8]; this spectrum distribution results from the diagonal or off-diagonal random of the metallic grain ensemble. Wigner’s theory predicts that the nearest-neighbor (NN) levels tend to avoid distribution too closely or too far; rather these spectra display some special statistics associated with the symmetry of systems, e.g. the NN level correlation and the two-level correlation behaviors. The distribution function of the NN levels of the grain ensemble is

$$P_n(\varepsilon) = c_n \left( \frac{\pi \varepsilon}{\delta_L} \right)^n e^{-\pi \varepsilon^2/(4\delta_L^2)},$$

(2)

where $c_n$ is the normalized coefficient, $n$ is the ensemble symmetry parameter. The average level spacing is $\delta_L = 4\varepsilon_p/3N$, with $\varepsilon_p$ the Fermi energy and $N$ total electron number. For itinerant FM, $n = 2$, corresponding to the unitary ensemble in which the time-reversal symmetry is broken due to the spin scattering or in magnetic field, and $c_n = \pi/2$; $P(\varepsilon)$ represents the possibility of two NN levels of the IE with separation $\varepsilon$ near the Fermi energy. And the two-level correlation function of the unitary ensemble is given by

$$R(\varepsilon - \varepsilon') = 1 - \sin^2 \left[ \frac{\pi(\varepsilon - \varepsilon')}{\delta_L} \right] \left[ \frac{\pi(\varepsilon - \varepsilon')}{\delta_L} \right],$$

(3)

where $\varepsilon$ and $\varepsilon'$ refer to two levels with arbitrary spacing in the grain ensemble.

Since we are interested in the effects of the spectral statistics on the magnetic properties, we employ the well-known mean-field or random-phase formulæ in the itinerant FM theory to study these effects. With the Hubbard model (1), the paramagnetic (PM) spin susceptibility is given by

$$\chi(0) = \frac{\chi_p}{1 - 2 U \chi_p/N} \quad \text{with}$$

$$\chi_p = \frac{1}{2} \sum_i \left\langle \frac{\partial \mathcal{F}}{\partial \varepsilon} \right\rangle_{\varepsilon = \mu},$$

(4)

where $\chi_p$ is the Pauli PM susceptibility of the IEs, $f = 1/(e^{\beta(\varepsilon - \mu)} + 1)$ denotes the Fermi–Dirac distribution function, $\langle \cdots \rangle$ means averaging over all the possible configurations of the metallic grain, or ensemble average. Hereinafter the susceptibility is measured with the units of $4\mu_B^2$. The summation in Eq. (4) over the energy level can become an integration over the energy in terms of the level distribution, as in Ref. [9]. One obtains the Pauli susceptibility for $N \gg 1$ at $T = 0$ K:

$$\chi_p = \begin{cases} \frac{R(\varepsilon_p)/(2\delta_L)}{1/(2\delta_L)} & \text{for odd grains,} \\ \frac{(2\pi - 0.5 - 0.5\pi e^{-4\pi})/8/(2\delta_L)}{0.72/(2\delta_L)} & \text{for even grains} \end{cases}$$

(5)

for the odd- and even-parity grains, respectively. From Eq. (5) one sees that the inverse Pauli PM susceptibility in odd or even grains is simply proportional to the average level spacing, $\delta_L$. Since $1/\delta_L$ approximately describes the density of states (DOS) near the Fermi level of the metallic nanostructures, with the electron number increasing to infinity ($N \to \infty$), $1/\delta_L$ is replaced by the DOS of infinite systems; therefore, in the limit the macroscopic result coincides with that of the macroscopic systems.

Additionally one obtains the spontaneous magnetization per site:

$$m^2 = \frac{M}{N\mu_B} = 2A \sum_i \left\langle \mathcal{F} \right\rangle_{\varepsilon = \mu},$$

(6)

where the spin exchange splitting $2A = U(\langle n_i \rangle - \langle n_i \rangle)$, $M$ is the total magnetization of the grain, $\mu_B$ is the occupancy of the electrons. The magnetization is in units of $\mu_B$. The ensemble average in $\langle \cdots \rangle$ over all of the configurations manifests the average over all of the possible level distributions. Without difficulty, one can obtain the zero-temperature spontaneous magnetization for $N \gg 1$:

$$m = \begin{cases} \frac{\Delta R(\varepsilon_p)/\delta_L}{A/\delta_L} \approx A/\delta_L & \text{for odd grains,} \\ 2A\langle n_i \rangle \approx 0.72A/\delta_L & \text{for even grains} \end{cases}$$

(7)

for the odd- and the even-parity metallic grains, respectively. So we find a profoundly simple result, that is, the spontaneous magnetization of the nanoscale metallic grains is proportional to the ratio of the spin exchange splitting over the average level spacing. Though there exists a slight difference between the odd and the even grains, the magnetization of the odd grain is larger than that of the even grain with almost the same level spacing and spin splitting. This result has a simple physical interpretation. In the FM grains when the spin splitting of the spin-up electrons with respect to the spin-down ones is $A$, and the average level spacing is $\delta_L$, the net number of up spin of IE is about $A/\delta_L$; therefore the magnetization is proportional to $A/\delta_L$.

3. The Stoner criterion

From the mean-field theory we know that the condition for the appearance of the FM long-range order, i.e.
the Stoner criterion in transition invariant metals is $2U_{F}(0)/N \geq 1$ [10,11]. This condition enforces that the spontaneous magnetization of the system is finite even if in the absence of magnetic field. Since the zero-temperature Pauli susceptibility is given by Eq. (5) for odd and even grains; thus, the Stoner criterion says that the Coulomb interaction $U$ must be greater than a critical value $U_c = N/2\delta_F(0)$, or

$$U_c = \begin{cases} N\delta_F R(e_F) \approx 1.33e_F & \text{for odd grains,} \\ N/(2\delta_F^{(\text{even})}(0)) \approx 1.85e_F & \text{for even grains,} \end{cases}$$

(8)

for the odd and the even grains, respectively. Compare the odd-parity grains with the even-parity grains which has almost the same Fermi energy, $U_{c}(\text{even}) > U_{c}(\text{odd})$, so the odd-parity grains can form FM long-range order more easily than the even grains do, which is similar to the formation of the local magnetic moment in nanoscale metallic grains [9].

This result implies that when the Coulomb interaction of metallic grains is smaller than the Fermi energy, the system will not exhibit FM long-range order. The physical meaning of the result could be understood as follows: the Coulomb interaction must be so large that the eigenvalue energy of the IE with the reversal spin is partially or fully pushed to the position above the Fermi energy; thus the system becomes spin polarized, net magnetic moment appears and the FM long-range order is established.

4. Spin wave dynamics

The itinerant FM in nanoscale systems is quasi-long-range order since the spin correlation is finite; this leads the spin wave excitation in nanoscale grains to be different from that in the translation-invariant systems. According to the standard random-phase-approximation result, the spin wave spectrum in itinerant metallic grains is given by the poles of the transverse propagator of the spins [12]. One gets the spin wave spectrum for the $s$th excitation, $\omega_s$, after average over all the possible configuration of the grain ensemble,

$$\hbar \omega_s = \tilde{\epsilon} - 2A + \frac{\pi U}{N} \sum_{n=1}^{\infty} \left( -1 \right)^n \frac{(nU/\delta_F)^{2n+1}}{(2n+1)!} \times \left[ n_p^{2n+1} - \left( n_p - \frac{\tilde{\epsilon}}{U} \right)^{2n+1} \right]$$

(9)

for the odd grains, where $\tilde{\epsilon}$ denotes the mean spacing of the NN levels in the two levels $\epsilon_i$ and $\epsilon_{i\pm s}$, $\tilde{\epsilon} = \langle \epsilon_{i\pm s} - \epsilon_i \rangle / s \sim \delta_L$. For the even grains, the spin wave spectrum is similar to Eq. (9) but with a more complicated expression in the last term.

Now we examine the behaviors of the spin wave excitation. To discuss the long-wavelength behavior at zero temperature, let $s = 0$, in the presence of the quasi-long-range order, $n_1 \neq n_1$, and the spin splitting $A$ is finite, the spin wave energy being

$$\hbar \omega(s = 0) \approx 2A(U/(N\delta_F) - 1)$$

$$= 2A(3U/4e_F - 1) \geq 0.$$  

(10)

Obviously, the spin wave energy (10) does not vanish even in the long-wavelength limit since the Stoner criterion holds, there exists a small gap for the spin wave excitation in nanoscale metallic FMs at $q \to 0$. This result is in contrast to the gapless spin wave spectrum in translation-invariant itinerant FMs. This gap is attributed to the broken translation invariance and the finite spin correlation length in nanoscale metallic grains. Also we find the $s$th spin wave excitation energy for small $s$ is about $\tilde{\epsilon}$ higher than the ground state due to $\delta_F/U \ll 1$, suggesting that the first few excitations are almost equi-spacing.

5. Conclusions

Experimentally, we would expect that the neutron scattering experiment on the nanoscale grains of the itinerant transition-metal Fe, Co or Ni embedded in nonmagnetic hosts may probe these unusual low-temperature magnetic properties.

In summary, the quantum level correlation and spectral statistics effect play crucial roles in the itinerant ferromagnetic properties and spin wave dynamics for the nanoscale metallic grains.

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