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Short-range order above the Curie temperature in the dynamic spin-fluctuation theory

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ABSTRACT

Based on the dynamic spin-fluctuation theory, we study the spin-density correlations in the ferromagnetic metals. We obtain computational formulae for the correlation function and correlation radius in different approximations of the theory. Using these formulae, we calculate the magnetic short-range order above the Curie temperature in bcc Fe. Results of the calculation confirm our theoretical prediction that the inverse correlation radius increases linearly with temperature for T sufficiently large. The calculated short-range order is small but sufficient to correctly describe neutron scattering experiments. A considerable amount of the short-range order is shown to persist up to temperatures much higher than the Curie temperature.

1. Introduction

The neutron scattering experiments in the ferromagnetic metals point on the existence of strong short-range order (SRO) above the Curie temperature T_C (see, e.g., [1–3]). However, the estimate of about 15–20 Å is based on the paramagnetic spin waves assumption, which has been strongly contested (see, e.g., [4,5]).

Theories of metallic magnetism [6–13] all postulate that local moments remain above T_C. But there is no agreement about the extent of the SRO in the paramagnetic phase. The fluctuating-local-band theory [6,7] is based upon the existence of very strong SRO well above T_C. However, it is difficult to test this approach by realistic calculations. The static spin-fluctuation theories [8,9] describe the paramagnetic phase as having no appreciable SRO outside the critical region. The situation is similar in various versions of the dynamic mean-field theory [10–12] and dynamical coherent potential approximation [13], as all of them build upon the single-site approximation.

The dynamic spin-fluctuation theory (DSFT) [14–16] takes into account both single-site and nonlocal interactions without mapping of the itinerant electron system onto an effective Hamiltonian with classical spins [17,12,18]. However, at high temperatures, the underlying Gaussian approximation (GA) leads to a discontinuous temperature dependence of magnetization, contrary to experiment.

In the extended DSFT [19], firstly the coupling of the spin fluctuations is improved by taking into account higher-order terms in the free energy. Secondly the nonlocal spin correlations are enhanced by taking into account uniform fluctuations in the single-site mean Green function. This combination of the renormalized Gaussian approximation (RGA) and enhanced uniform fluctuations (UF) makes the extended DSFT applicable at all temperatures. In particular, the extended theory eliminates the fictitious first-order phase transition and yields a proper second-order phase transition from the ferromagnetic to paramagnetic state.

In this paper we apply the extended DSFT to study SRO in the paramagnetic state. For the spin correlator in the extended DSFT, we derive an explicit expression and approximate formula of the Ornstein–Zernike type. As a measure of the SRO, we use the halfwidth of the normalized spin correlator and correlation radius

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of the Ornstein–Zernike type correlator. The inverse correlation radius is shown to change linearly with temperature in close analogy to the Curie–Weiss law.

Results in the RGA+UF are illustrated in the example of bcc Fe. We calculate the spin correlator as a function of distance and temperature. For selected temperature values, the spin correlator as a function of distance was calculated in the static [20] and dynamic [21,14] approximations of the SFT but no definite conclusions about the extent of the SRO in Fe were drawn. We obtain a quantitative estimate of the SRO and give an interpretation of the neutron scattering experiments based on calculations of the correlation halfwidth, correlation radius and their inverse as functions of temperature.

The paper is organized as follows. In Section 2, we obtain computational formulae for the correlation function and correlation radius in different approximations of the DSFT. Using these formulae, in Section 3, we calculate SRO above the Curie temperature in bcc Fe and compare the results with existing theoretical work and neutron scattering experiments. In Section 4, we summarize the present work.

2. Spatial correlator

We study the spatial correlator $\langle s^\alpha(\mathbf{r})s^\alpha\rangle$ of the spin-density operator $s^\alpha(\mathbf{r})$, $\alpha = x, y, z$. Here, the angle brackets denote the canonical average $\langle\ldots\rangle = Z^{-1}\,\mathrm{Tr}(\ldots \mathrm{e}^{-H/T})$, where $Z = \mathrm{Tr}\,\mathrm{e}^{-H/T}$ is the partition function, H is the Hamiltonian of the system of interacting electrons, and T is temperature (in energy units). Since we consider the spatial correlator in the *paramagnetic* region, the average $\langle s^\alpha(\mathbf{r})s^\alpha\rangle$ does not depend on the index α. For brevity, we omit the index α further on.

The spatial Fourier transformation is defined by

$$s_{\mathbf{q}} = \int s(\mathbf{r})\mathrm{e}^{-\mathrm{i}\mathbf{q}\mathbf{r}}\,\mathrm{d}\mathbf{r}, \quad s(\mathbf{r}) = \frac{1}{N_\mathrm{a}\Omega_\mathrm{WS}}\sum_{\mathbf{q}} s_{\mathbf{q}}\,\mathrm{e}^{\mathrm{i}\mathbf{q}\mathbf{r}},$$

where N_a is the number of atoms, Ω_WS is the volume of the Wigner–Seitz cell and the integral is taken over the whole volume of the crystal. The translational invariance of the system leads to

$$\langle s(\mathbf{r})s\rangle = \frac{1}{N_\mathrm{a}^2\Omega_\mathrm{WS}^2}\sum_{\mathbf{q}}\langle s_{\mathbf{q}}s_{-\mathbf{q}}\rangle\,\mathrm{e}^{\mathrm{i}\mathbf{q}\mathbf{r}}. \tag{1}$$

Now we derive an expression for the spin correlator in the momentum representation, $\langle s_{\mathbf{q}}s_{-\mathbf{q}}\rangle$. The thermodynamic susceptibility $\chi_{\mathbf{q}m}$, in the units of $\frac{1}{2}g^2\mu_\mathrm{B}^2$ (g is the electron g-factor, and μ_B is the Bohr magneton), is related to the spin correlator

$$\langle s_{\mathbf{q}m}s_{-\mathbf{q}-m}\rangle = T\int_0^{1/T}\langle s_{\mathbf{q}}(\tau)s_{-\mathbf{q}}\rangle\,\mathrm{e}^{\mathrm{i}\omega_m\tau}\,\mathrm{d}\tau$$

by the formula [22]

$$\chi_{\mathbf{q}m} = \frac{2}{T}\langle s_{\mathbf{q}m}s_{-\mathbf{q}-m}\rangle.$$

Here $s_{\mathbf{q}}(\tau) = \mathrm{e}^{\tau H}s_{\mathbf{q}}\,\mathrm{e}^{-\tau H}$ is the 'Heisenberg' representation with respect to the 'time' $\tau \in [0, 1/T]$ and $\omega_m = 2\pi mT$ are even thermodynamic 'frequencies'. Applying the inverse Fourier transformation with respect to 'time', at $\tau=0$ we have

$$\langle s_{\mathbf{q}}s_{-\mathbf{q}}\rangle = \sum_m\langle s_{\mathbf{q}m}s_{-\mathbf{q}-m}\rangle = \frac{T}{2}\sum_m\chi_{\mathbf{q}m}.$$

Replacing the sum over even 'frequencies' ω_m by an integral with the Bose function $B(\varepsilon) = (\mathrm{e}^{\varepsilon/T}-1)^{-1}$ and neglecting the zero-point fluctuations, which lead to an insignificant shift of the self-energy [19], we come to

$$\langle s_{\mathbf{q}}s_{-\mathbf{q}}\rangle = \frac{1}{2\pi}\int B(\varepsilon)\mathrm{Im}\,\chi_{\mathbf{q}}(\varepsilon)\mathrm{d}\varepsilon \tag{2}$$

in full agreement with the fluctuation–dissipation theorem. For further calculations, we need to obtain an explicit expression for $\chi_{\mathbf{q}}(\varepsilon)$.

In the DSFT the enhanced susceptibility $\chi_{\mathbf{q}}(\varepsilon)$ is expressed in terms of the unenhanced one $\chi^0_{\mathbf{q}}(\varepsilon)$ by the formula

$$\chi_{\mathbf{q}}(\varepsilon) = \frac{\chi^0_{\mathbf{q}}(\varepsilon)}{1 - u\chi^0_{\mathbf{q}}(\varepsilon)}, \tag{3}$$

where u is the effective interaction constant. Using formula (3), we write expression (2) as

$$\langle s_{\mathbf{q}}s_{-\mathbf{q}}\rangle = \frac{1}{\pi u}\int_0^\infty B(\varepsilon)\mathrm{Im}\,\frac{1}{1 - u\chi^0_{\mathbf{q}}(\varepsilon)}\,\mathrm{d}\varepsilon. \tag{4}$$

Because of strong localization of the Bose function at zero energy, it suffices to know the behavior of the susceptibility only for small ε. We expand the susceptibility in Taylor series

$$\chi^0_{\mathbf{q}}(\varepsilon) = \chi^0_{\mathbf{q}}(0) + \mathrm{i}\frac{d\chi^0_{\mathbf{q}}(0)}{d\varepsilon}\varepsilon \equiv \chi^0_{\mathbf{q}}(0) + \mathrm{i}\varphi_{\mathbf{q}}\varepsilon$$

(taking into account that $\mathrm{Im}\,\chi^0_{\mathbf{q}}(\varepsilon)$ is an odd function and $\mathrm{Re}\,\chi^0_{\mathbf{q}}(\varepsilon)$ is an even one) and use the following approximation of the Bose function:

$$\frac{1}{\mathrm{e}^{\varepsilon/T}-1} \approx \begin{cases} T/\varepsilon, & \varepsilon < \varepsilon_0, \\ 0, & \varepsilon > \varepsilon_0, \end{cases}$$

where the value $\varepsilon_0 = \pi^2T/6$ is chosen such that the first moments are equal. As a result, formula (4) takes the form

$$\langle s_{\mathbf{q}}s_{-\mathbf{q}}\rangle = \frac{T}{2u\lambda_{\mathbf{q}}}\frac{2}{\pi}\arctan\frac{u\varphi_{\mathbf{q}}\pi^2T}{6\lambda_{\mathbf{q}}}, \tag{5}$$

where $\lambda_{\mathbf{q}} = 1 - u\chi^0_{\mathbf{q}}(0)$.

The interaction of the modes is taken into account by interpolating the static susceptibility $\chi^0_{\mathbf{q}}$ between the homogeneous susceptibility χ^0_0 and the local susceptibility $\chi^0_\mathrm{L} = N_\mathrm{a}^{-1}\sum_{\mathbf{q}}\chi^0_{\mathbf{q}}$:

$$\chi^0_{\mathbf{q}} = \chi^0_0 + (\chi^0_\mathrm{L} - \chi^0_0)q^2/\overline{q^2}, \tag{6}$$

where $\overline{q^2}$ is the average of q^2 over the Brillouin zone. The local susceptibility χ^0_L is determined by the single-site mean Green function $g(\varepsilon)$ (for detailed formulas, see [15]).

To improve the temperature dependence of magnetization at high temperatures, in the extended DSFT [19] the coupling of the spin fluctuations is improved by taking into account higher-order terms of the free energy in the fluctuating field. The use of the decoupling techniques leads to a renormalization of the mean field $\bar{V}_z$ due to the third-order terms:

$$\bar{V}_z^\mathrm{RGA} = (1+\eta)\bar{V}_z^\mathrm{GA},$$

and to renormalization of the susceptibility due to the fourth-order terms:

$$\chi^\mathrm{RGA}_{\mathbf{q}}(\varepsilon) = (1+3\eta)\chi^\mathrm{GA}_{\mathbf{q}}(\varepsilon).$$

In the paramagnetic region, the correction coefficient η is calculated self-consistently as

$$\eta = -\frac{3\pi}{WN_\mathrm{d}}\chi_\mathrm{L}^\mathrm{GA}(0)\langle\Delta V^2\rangle',$$

where W is the bandwidth, $N_\mathrm{d} = 5$ is the number of degenerate d bands, and $\langle\Delta V^2\rangle'$ is the local thermal fluctuation net of intrinsic field fluctuations. Another feature of the extended DSFT is the