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A single-site spin correlation function in paramagnetic iron

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Abstract. A single-site dynamic spin correlation function in paramagnetic iron is considered, taking into account electron–electron correlation effects in the single-site approximation of multiple scattering theory. Correlation effects significantly change the spin correlation function, especially at low frequencies, and have allowed us to obtain, in the energy interval $0 \leq \hbar\omega \leq 0.1$ eV, the value $1.3 \mu_B$ of the local magnetic moment, in accordance with polarized neutron scattering experiments. The damping time for the spin correlation function is estimated to be approximately 10^{-13} s, which greatly exceeds the electron hopping time, $\sim 10^{-15}$ s.

1. Introduction

The widely discussed question of the character of spin excitations in itinerant electron systems of metallic magnets (see e.g. Panel discussion on ICM-82 1983) and also problems arising from interpretation of experiments on neutron scattering (Lynn and Mook 1981, Wicksted *et al* 1984, Mook 1988) pose the problem of the calculation of the dynamic spin correlation function. The spin correlation function in magnetic metals has been considered in both theoretical (Edwards 1982, 1983, Capellmann and Vieira 1982, Oguchi *et al* 1983, Hasegawa 1983, Gyorffy *et al* 1985) and experimental works (Brown *et al* 1982, Ishikawa 1983, Shirane *et al* 1986). Most of these works dealt with the space variation of spin density, which determines the short-range magnetic order. Of similar importance is the time dependence of the electron spin density, because in experiments on neutron and electron scattering, photoemission, etc, it is necessary to take into account the relation between the measurement time, i.e. the time of interaction of an external force with the system, and the relaxation time. However, to date the problem of the time dependence of spin excitations in an electron system has not been studied in detail, although the problem was discussed on a qualitative level by Capellmann (1982) and Edwards (1984).

Over the last decade increasing interest in different fluctuation theories of transition-metal magnetism has been observed (see e.g. Moriya 1986, Capellmann 1987). These theories clearly lead to the conclusion that the fluctuations of electron spin density play an important, and sometimes determining, role in many properties of transition metals. In the construction of these theories it is also important to make clear the relation between the time for formation of an electron state in metals (which is comparable with

the time for electron transfer from one site to a neighbouring site in the crystal lattice) and the time for formation and changing of local magnetic moments. In particular, if the time of magnetic moment variation τ_m is greater than the time of electron hopping $\tau_e \sim \hbar/W \sim 0.1\text{--}1$ fs (W is the bandwidth and 1 fs = 10^{-15} s), then one may use, for the description of the electron scattering mechanism, an approximation analogous to the adiabatic approximation in the crystal lattice theory, and thus substantiate, in a sense, the widely used static approximation for spin fluctuation theories.

The spin correlation function is connected with the dynamic susceptibility by the fluctuation-dissipation theorem. Therefore studies of susceptibility may help to construct the time dependence of local magnetic moments. One of the first and consistent works on calculations of the dynamic susceptibility in metallic systems was the work by Cooke *et al* (1980). The dynamic susceptibility and also spin-wave energies for iron and nickel calculated there are in good agreement with experiment. Analogous calculations were carried out by Callaway *et al* (1983). But all these calculations are applicable to metallic magnets only at relatively low temperatures, since the random-phase approximation (RPA) used in these works describes only long-wave magnons well enough. At temperatures comparable with the phase transition temperature (for strong ferromagnets $T_C \sim 1000$ K) the short-wave excitations (spin fluctuations) become important. Therefore it is necessary to modify the calculation scheme so that it accounts for short-wave excitations. One of the simplest variants of such accounting will be considered in this paper.

2. Local spin susceptibility

The Fourier transform of the spin susceptibility is given by

$$\chi_{nr'}^{\alpha\beta}(\omega) = \frac{i}{\hbar} \int_0^\infty \langle [\hat{S}_n^\alpha(t), \hat{S}_n^\beta(0)] \rangle e^{i\omega t - \delta t} dt. \quad (2.1)$$

Here $\hat{S}_n^\alpha(t)$ is the spin moment operator on the n th site of the lattice, taken in the Heisenberg representation with total Hamiltonian \hat{H} . Index α ($= x, y, z$) specifies components of the spin operator. Angle brackets in (2.1) denote quantum statistical averaging and square brackets denote commutation. An infinitesimal positive addition δ provides convergence of the corresponding integrals. The magnetic moment operator is connected with the spin moment operator by the relation $\hat{M} = -g\mu_B \hat{S}$.

The site spin moment operator is determined via the operator of electron spin density $\hat{S}(r, t)$:

$$\hat{S}_n(t) = \int_{\Omega_n} \hat{S}(r, t) dr \quad (2.2)$$

where Ω_n is the volume of the Wigner-Seitz cell centred on the n th site. The electron spin density is formed by spin moments \hat{s}_i of all electrons:

$$\hat{S}(r, t) = \sum_i \hat{s}_i(t) \delta(r - r_i(t)). \quad (2.3)$$

For systems described by a one-electron Hamiltonian

$$\hat{H}_0 = \sum_i \varepsilon_i a_i^\dagger a_i \quad (2.4)$$

the susceptibility may be obtained as

$$\chi_{nn'}^{0,\alpha\beta}(\omega) = \frac{1}{4} \sum_{l'l'} \frac{e^{i(k-k')R_{nn'}} |J_l^{\alpha'}(n')|^2 \tau_{\sigma'\sigma}^{\alpha} \tau_{\sigma\sigma'}^{\beta}}{\varepsilon_{l'} - \varepsilon_l + \hbar\omega + i\hbar\delta} (f_l - f_{l'}). \quad (2.5)$$

Here ε_l is the one-electron energy, a_l (a_l^\dagger) is an annihilation (creation) operator of an electron in state l in the second quantized representation. Index $l = (k, \lambda, \sigma)$ involves the electron wavevector k , the band number λ and the spin projection index $\sigma = \pm 1$ (\uparrow, \downarrow). Vector R_n connects the origin of coordinates with the n th site of the lattice, and $R_{nn'} = R_n - R_{n'}$. The equation (2.5) includes an integral

$$J_l^{\alpha'}(n) \equiv J_{k\lambda\sigma}^{k'\lambda'\sigma'}(n) = \int_{\Omega_n} \varphi_{k'\lambda'\sigma'}^*(\mathbf{r}) \varphi_{k\lambda\sigma}(\mathbf{r}) d\mathbf{r} \quad (2.6)$$

where $\varphi_{k\lambda\sigma}(\mathbf{r})$ is the one-electron Bloch function normalized to unity over all the crystal. Besides that, the formula (2.5) includes matrix elements $\tau_{\sigma\sigma'}^{\alpha}$ of the Pauli matrices τ^x, τ^y, τ^z and the Fermi-Dirac distribution function $f_l = \{1 + \exp[(\varepsilon_l - \mu)/k_B T]\}^{-1}$, μ being the chemical potential.

The problem of susceptibility calculations for a system of interacting electrons is more difficult, because it is necessary to take into account many-particle effects of electron correlations. At this point, the susceptibility of the system may differ significantly from the 'zero' susceptibility of a system of non-interacting electrons, described by the one-electron Hamiltonian (2.4). Let us suppose that the total Hamiltonian \hat{H} of the electron system has Hubbard's form:

$$\hat{H} = \hat{H}_0 + I \sum_n \hat{\rho}_{n\uparrow} \hat{\rho}_{n\downarrow} \quad (2.7)$$

where $\hat{\rho}_{n\sigma}$ is the electron number operator on the n th site with spin projection σ and I is the effective Stoner-Hubbard constant of the electron-electron interaction. Henceforward, we are considering the paramagnetic case, assuming all directions in space to be equivalent.

In the standard RPA the total susceptibility can be expressed through the zero susceptibility (Izuyama *et al* 1963):

$$\chi(\mathbf{q}, \omega) = \chi^0(\mathbf{q}, \omega) / [1 - 2I\chi^0(\mathbf{q}, \omega)] \quad (2.8)$$

where \mathbf{q} is the wavevector. In site representation, formula (2.8) has the form

$$\chi_{nn'}^{\sigma\sigma'}(\omega) = \chi_{nn'}^{0,\sigma\sigma'}(\omega) + \sum_{n''\sigma''} \chi_{nn''}^{0,\sigma\sigma''}(\omega) I \chi_{n''n'}^{\sigma''\sigma'}(\omega). \quad (2.9)$$

However, formula (2.8) inadequately describes susceptibility at high temperatures, because in obtaining it the interaction between different modes of spin excitations was neglected, which is, in some sense, equivalent to a long-wave approximation.

The more general formula for inter-site susceptibility is

$$\chi_{nn'}^{\sigma\sigma'}(\omega) = \chi_{nn'}^{0,\sigma\sigma'}(\omega) + \sum_{n''\sigma''} \chi_{nn''}^{0,\sigma\sigma''}(\omega) \Gamma_{n''}(\omega) \chi_{n''n'}^{\sigma''\sigma'}(\omega). \quad (2.10)$$

Here $\Gamma_n(\omega)$ is the effective vertex part, which describes effects of multiple electron scattering. In deriving formula (2.9) it is supposed that all sites are equivalent and electron-electron interaction is the same on all sites and determined by constant I . As for formula (2.10), it takes into account the difference of electron interaction for different energies and wavelengths. This difference is connected with the existence of thermal and

quantum excitations of electron density, which lead to an inhomogeneous distribution of potentials in both space and time.

At high temperatures the electron system is strongly inhomogeneous because of large fluctuations of spin and charge density. This leads to almost complete loss of memory about electron wave phase on electron transition from one site to another. As a result, through multiple scattering on fluctuation potentials of neighbouring sites, the electrons arrive at a given site with different phases, and contributions to the susceptibility from inter-site transitions are extinguished. In this case it is sufficient in the sum of equation (2.10) to retain only one term with $n = n'$. We will call this approach the single-site approximation (SSA). Neglecting the frequency dependence of the function Γ_n , we replace Γ_n by an effective constant U . Then for paramagnetic local susceptibility

$$\chi_L \equiv \chi_{nn}^{zz} = \frac{1}{4}(\chi_{nn}^{\uparrow\uparrow} + \chi_{nn}^{\downarrow\downarrow} - \chi_{nn}^{\uparrow\downarrow} - \chi_{nn}^{\downarrow\uparrow}) \quad (2.11)$$

we have

$$\chi_L^{\text{SSA}}(\omega) = \chi_L^0(\omega)/[1 - 2U\chi_L^0(\omega)] \quad (2.12)$$

where $\chi_L^0(\omega)$ is the zero local susceptibility obtained from (2.5) at $\alpha = \beta$ and $n = n'$.

The usual RPA susceptibility (2.8) contains enhancement for one fixed q -mode only. This approach is suitable for an ideal crystal. But at high temperatures ideality is broken and the strong interaction of modes with different q takes place. The simplest way to account for this circumstance is to replace $\chi^0(q, \omega)$ in the denominator of (2.8) by an average:

$$\chi_L^0(\omega) \equiv \frac{1}{N} \sum_q \chi^0(q, \omega) \quad (2.13)$$

N being the number of q -vectors in the Brillouin zone. Then, after summation over q , it is obtained from (2.8) that

$$\chi_L(\omega) = \chi_L^0(\omega)/[1 - 2I\chi_L^0(\omega)] \quad (2.14)$$

which is equivalent to exclusion of all terms in (2.9) with $n \neq n'$. Although formula (2.12) coincides in structure with formula (2.14), it includes the parameter U , which is the renormalized constant I with due regard for multiple electron scattering on spin and charge fluctuations.

At high temperatures the local spin susceptibility $\chi_L^{\text{SSA}}(\omega)$ is preferable to

$$\chi_L^{\text{RPA}}(\omega) = \frac{1}{N} \sum_q \chi(q, \omega)$$

since the main contribution to thermodynamic quantities in this case is due to short-wave spin excitations.

In this work we concentrate on the spin correlation function, for which the method of polarized neutron scattering remains the main experimental method. Characteristic neutron excitation energies are dozens of millielectronvolts (meV), which is much less than the energy of Stoner excitations. For this reason we are especially interested in the low-frequency region.

At low frequencies the imaginary part of the zero local susceptibility is linear in ω , which is easily shown by replacing the matrix elements $J_i^{\alpha\beta}$ in formula (2.5) by a constant

J. Then the imaginary part of the zero susceptibility $\text{Im } \chi_L^0(\omega)$ may be expressed analytically in terms of the density of states (DOS) $\nu(\varepsilon)$:

$$\text{Im } \chi_L^0(\omega) = \frac{\pi}{2} J^2 \int_{-\infty}^{\infty} f(\varepsilon) \nu(\varepsilon) [\nu(\varepsilon + \hbar\omega) - \nu(\varepsilon - \hbar\omega)] d\varepsilon. \quad (2.15)$$

For low frequencies this formula takes the form

$$\text{Im } \chi_L^0(\omega) = \frac{\pi}{2} J^2 \hbar\omega \int_{-\infty}^{\infty} \left(-\frac{\partial f}{\partial \varepsilon} \right) \nu^2(\varepsilon) d\varepsilon. \quad (2.16)$$

Finally, neglecting thermal smearing of the Fermi-Dirac function, we get a simple formula

$$\text{Im } \chi_L^0(\omega) = \frac{1}{2} \pi J^2 \nu^2(\varepsilon_F) \hbar\omega \equiv \gamma \hbar\omega. \quad (2.17)$$

Thus, the linear behaviour of the imaginary part of the zero susceptibility at low frequencies is generally determined by the DOS on the Fermi level ε_F . Formula (2.17) can be used for estimation of the ω dependence of the zero susceptibility at low frequencies for various transition metals.

The real part of the zero local susceptibility in the upper half-plane is connected with the imaginary part by the Hilbert transformation

$$\text{Re } \chi_L^0(\omega) = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\text{Im } \chi_L^0(\omega')}{\omega' - \omega} d\omega'. \quad (2.18)$$

This allows us to reconstruct $\text{Re } \chi_L^0$ from the known imaginary part, $\text{Im } \chi_L^0$. The real part, $\text{Re } \chi_L^0(\omega + i0)$, as a function of ω has the usual behaviour. At high frequencies $\text{Re } \chi_L^0(\omega) \sim -1/\omega^2$ because of the oddness of $\text{Im } \chi_L^0(\omega)$. In the intermediate region the function $\text{Re } \chi_L^0(\omega)$ changes its sign. At zero frequency $\text{Re } \chi_L^0$ has a maximum. Especially important for further analysis is a peak-like behaviour of the real part of the zero local susceptibility at low frequencies. Just this singularity leads to the singularity in the total local susceptibility.

Essential changes in the susceptibility behaviour are induced by many-particle correlation effects, which may greatly increase susceptibility in the low-frequency region. Indeed, let us write the imaginary part of the total local susceptibility (2.12) as

$$\text{Im } \chi_L(\omega) = \frac{\text{Im } \chi_L^0(\omega)}{[1 - 2U \text{Re } \chi_L^0(\omega)]^2 + [2U \text{Im } \chi_L^0(\omega)]^2}. \quad (2.19)$$

Many-particle effects here are accounted for by the electron interaction constant U . The denominator in (2.19) has a resonance character at $U = 1/[2 \text{Re } \chi_L^0(\omega)]$, which, with a suitably chosen constant U , can give a radical increase in $\text{Im } \chi_L^0(\omega)$. Notice that the criterion for local magnetic moment formation has the form

$$U \geq U_c = 1/[2 \text{Re } \chi_L^0(0)]. \quad (2.20)$$

(On the criterion of local magnetism see, for example, Heine *et al* (1981).) Calculations by formula (2.19) for systems with $U \geq U_c$ will be incorrect, because at zero frequency these systems are unstable relative to creation of local magnetic moments. From this point of view, the calculation scheme of the ground state of an electron system must be changed so that local magnetic moments are taken into account. It is obvious that a full solution of this problem can be based on knowledge of the free energy of a system with

values of local magnetic moments variable in space and time. This consideration is in progress and will be published elsewhere.

Increasing the interaction constant U results in growth of the imaginary part of the susceptibility at low frequencies, the amplification of $\text{Im } \chi_L^0(\omega)$ taking on resonance character. Similar dependence of the paramagnetic susceptibility $\chi(q, \omega)$ on I in the RPA was noticed by Doniach (1967). The difference between the present work and Doniach's is that we consider total *local* susceptibility and carry out our 'zero' calculations with real electron energy bands, rather than with free electrons as in Doniach's work. The change of $\text{Im } \chi_L(\omega)$ with the increase of the constant U is connected with the 'pumping over' of intensity of the function $\text{Im } \chi_L(\omega)$ from the high-frequency to the low-frequency region. It is easy to prove that the local susceptibility satisfies the moments rule:

$$\int_0^\infty \omega \text{Im } \chi_L^0(\omega) d\omega = \int_0^\infty \omega \text{Im } \chi_L(\omega) d\omega. \quad (2.21)$$

So, electron correlation effects can greatly change the imaginary part of the zero susceptibility.

3. Single-site spin correlation function and local moments

The spin correlation function is determined as

$$A_{nn}^{\alpha\beta}(t) = \frac{1}{2} \langle \{ \hat{S}_n^\alpha(t), \hat{S}_n^\beta(0) \} \rangle. \quad (3.1)$$

The frequency correlation function is determined, as usual, by a Fourier transform:

$$A_{nn}^{\alpha\beta}(\omega) = \int_{-\infty}^{\infty} A_{nn}^{\alpha\beta}(t) e^{i\omega t} dt. \quad (3.2)$$

The fluctuation-dissipation theorem of Callen and Welton connects the correlation function (3.2) with the susceptibility (see e.g. White 1983):

$$A_{nn}^{\alpha\beta}(\omega) = \frac{\hbar}{2i} \coth\left(\frac{\hbar\omega}{2k_B T}\right) [\chi_{nn}^{\alpha\beta}(\omega) - \chi_{nn}^{*\beta\alpha}(\omega)]. \quad (3.3)$$

Here T is temperature, k_B is the Boltzmann constant and an asterisk means complex conjugate.

In the paramagnetic case, from (3.3) for the single-site frequency correlation function $A(\omega)$ we obtain

$$A(\omega) \equiv \sum_{\alpha} A_{nn}^{\alpha\alpha}(\omega) = 3\hbar \coth\left(\frac{\hbar\omega}{2k_B T}\right) \text{Im } \chi_L(\omega). \quad (3.4)$$

At high frequencies ($\omega \rightarrow \infty$) the multiplier $\coth(\hbar\omega/2k_B T)$ tends to unity and the function $A(\omega)$ is congruent with the $\text{Im } \chi_L(\omega)$ function. But at low frequencies $\coth(\hbar\omega/2k_B T) \approx 2k_B T/\hbar\omega$ and taking into account (2.19) and (2.17), we have

$$A(\omega) = 6\hbar \frac{\gamma k_B T}{[1 - 2U \text{Re } \chi_L^0(0)]^2 + (2U\gamma\hbar\omega)^2}. \quad (3.5)$$

As seen from this formula, the resonance denominator may greatly enhance the correlation function and give a peak in $A(\omega)$.

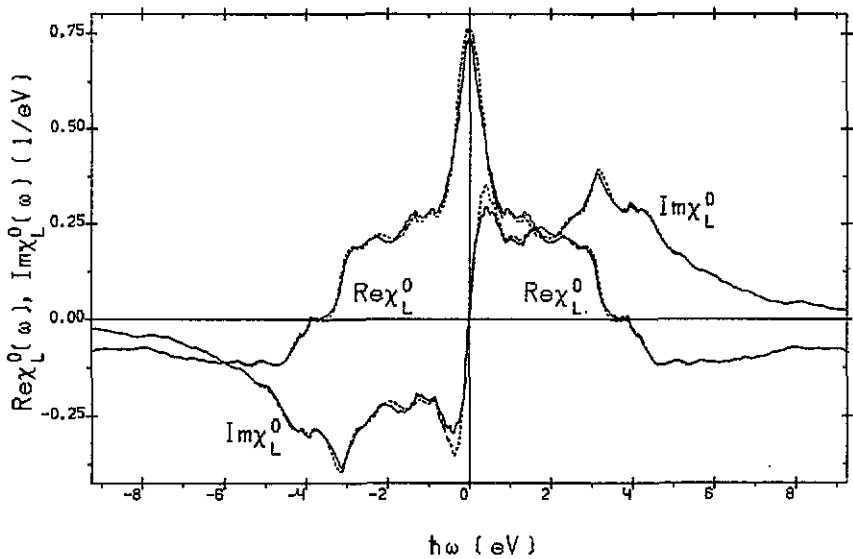


Figure 1. Real and imaginary parts of the zero local spin susceptibility at temperatures $T = 0$ K (broken curves) and $T = 1.25 T_C$ (full curves).

One of the general characteristics measured by polarized neutron scattering is the local spin moment magnitude. We determine the magnitude of an effective local spin moment in a frequency interval $[0, \omega]$ by the formula

$$S_L(\omega) = \left(\frac{1}{\pi} \int_0^\omega A(\omega') d\omega' \right)^{1/2}. \quad (3.6)$$

It is clear that in the infinite frequency interval this magnitude coincides with a root mean square of the local spin moment

$$S_L \equiv S_L(\infty) = \langle \hat{S}^2(t=0) \rangle^{1/2}. \quad (3.7)$$

4. Results of calculations

Calculations of energy bands $\varepsilon_{k\lambda\sigma}$ and wavefunctions $\varphi_{k\lambda\sigma}(r)$ for the ground state of paramagnetic iron were carried out by the standard Korringa-Kohn-Rostoker (KKR) method with a self-consistent crystal potential taken from the Moruzzi *et al* (1978). The zero local susceptibility was computed according to formula (2.5) with $\alpha = \beta$ and $n = n'$. The six-dimensional Brillouin zone integration in (2.5) was carried out by the tetrahedron method. Results of calculations of zero local susceptibility (real and imaginary parts) for paramagnetic iron at temperatures $T = 0$ and 1305 K ($1.25 T_C$) are given in figure 1. The characteristic peaks of the imaginary part of the susceptibility, $\text{Im} \chi_L^0(\omega)$, are due to electron DOS singularities and reflect the peculiarities of high-energy one-particle (Stoner) excitations, arising from intra- and inter-band transitions. It is seen that the imaginary part of the zero susceptibility has the first peak at a frequency $\hbar\omega = 0.5$ eV. It follows from this that considerable changes in zero local susceptibility and local magnetic moments due to one-particle excitations are beginning at frequencies

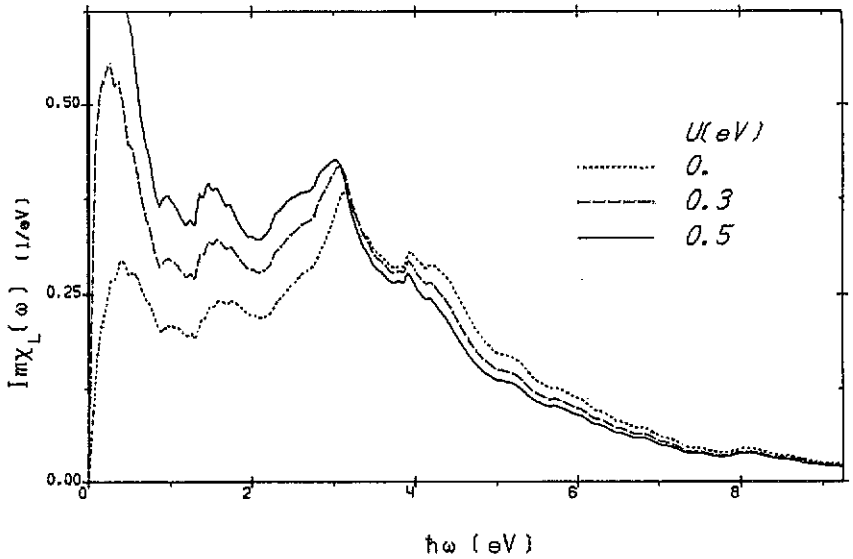


Figure 2. Imaginary part of the total local spin susceptibility.

higher than the frequency ω_C corresponding to the Curie point, $\hbar\omega_C = k_B T_C \approx 0.1$ eV. This result accords with the well known conclusion of the one-particle (Stoner) theory, which gives for the Curie point of iron the value ~ 4000 K, approximately four times higher than the experimental value $T_C = 1044$ K. Note also that one-particle excitations may be directly observed with the help of new rapidly developing experimental photo-emission methods, which allow one to look deep into an energy spectrum and to measure spectra of Stoner excitations (see e.g. Penn 1987 and references therein). As is obvious from figure 1, the temperature changes the zero susceptibility, but this change is small and cannot lead to significant alteration of results for the correlation function, calculated with zero susceptibility at $T = 0$ K.

In order to elucidate the role of the electron correlation effects, the total local susceptibility $\chi_L(\omega)$ and the spin correlation function $A(\omega)$ have been calculated for the values $U = 0, 0.3$ and 0.5 eV and for $T = 1.25 T_C$. In figure 2, plots of the imaginary part of the total susceptibility are given. In accordance with the moments rule (2.21), a great enhancement of the imaginary part of the susceptibility in the low-frequency region with increase of the constant U is observed. Peculiarities of the local susceptibility are conserved in this enhancement, and become more clear. The position of the low-energy peak of the imaginary part of the susceptibility curve moves to zero frequency with increasing U . In the high-energy region for $\hbar\omega > 3$ eV the deviations of $\text{Im } \chi_L$ curves from the $\text{Im } \chi_L^0$ curve are small. The same behaviour has been observed for the spin correlation function. In figure 3 the graphs of the spin correlation function $A(\omega)$ in the energy interval $[0, 0.8$ eV] are presented. As seen from the figure, the spin correlation function is greatly enhanced at low energies with increase of U . According to the formula (3.4) the enhancement of the spin correlation function is connected not only with the enhancement of the imaginary part of the susceptibility, but also with thermal enhancement through the factor $\coth(\hbar\omega/2k_B T)$. At energies $\hbar\omega > 2k_B T$ this factor is near unity and does not give a contribution to the correlation function.

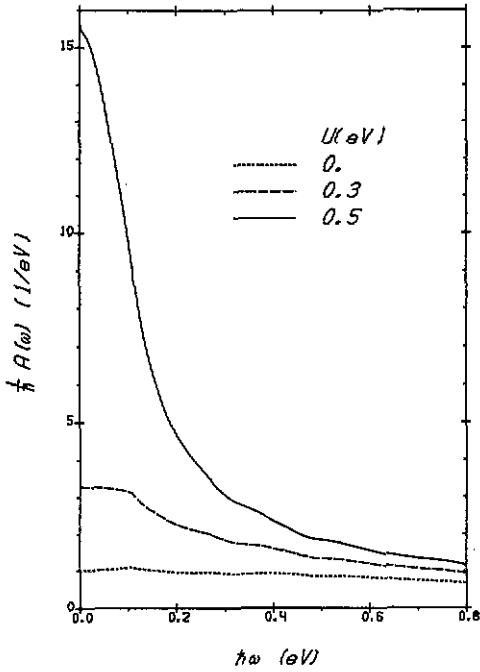


Figure 3. Frequency-dependent spin correlation function.

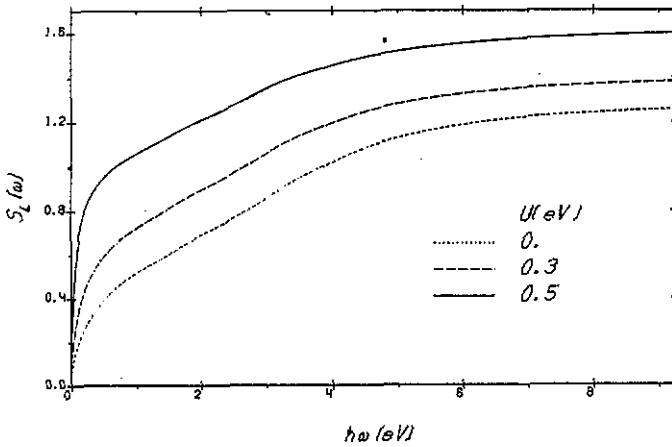


Figure 4. The local spin moment.

In figure 4 graphs of the magnitude of the effective local spin moment in paramagnetic iron are given as a function of interval length $[0, \omega]$ (see (3.6)) for different values of constant U . Comparing these graphs, it is not difficult to conclude that correlation effects essentially change the magnitude of the effective local moment, the changes amounting to dozens of times at some values of U and interval length. Measurements of local magnetic moments by polarized neutrons are made in a narrow energy window $\Delta = 0.1$ eV. Even in this window it is gathering a significant quantity of the local magnetic

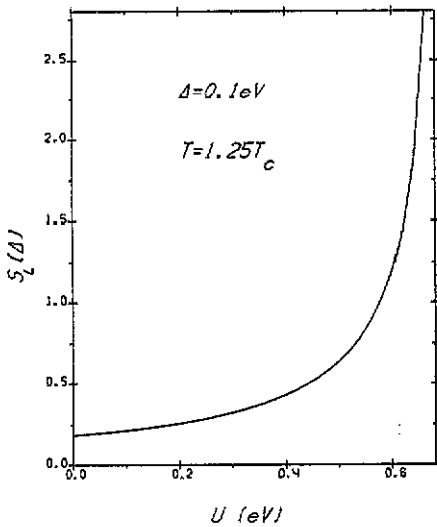


Figure 5. The local spin moment in the energy interval $\Delta = 0.1$ eV.

moment for iron: $M_L(\Delta) = 1.3 \mu_B$ ($S_L(\Delta) = 0.65$) (Brown *et al* 1982, Shirane *et al* 1986). But determination of local magnetic moments by polarized neutron scattering has a shortcoming, connected with the finite interaction time $\tau_n \sim 10^{-13}$ s between a neutron and a magnetic atom. It is clear that the high-frequency components of local moment with times $t \ll \tau_n$ give zero contribution to the cross section of polarized neutron scattering. Neutrons completely 'notice' only the low-frequency components of local magnetic moments with $t \gg \tau_n$. In the intermediate frequency region the components of local moments with times $t \sim \tau_n$ give only a partial contribution to the magnetic cross section. This shortcoming may be overcome by using an experimental technique with smaller interaction time than in conventional neutron experiments. For example, one could use polarized x-ray experiments or neutron experiments with large neutron energy, as in the work of Shirane *et al* (1986). For this reason, comparison of thermal neutron experiments with theoretical calculations should be carried out for large times $t > \tau_n$, which corresponds to low energies ≤ 0.1 eV. Thus, the correlation function, which is responsible for collecting the local magnetic moment, must contain major intensity in a narrow interval $\hbar\omega \leq 0.1$ eV. As we have seen above, this amplification of the correlation function may be connected only with many-particle effects.

In figure 5 the U dependence of the effective local spin moment is given for the energy window $\Delta = 0.1$ eV. The value of local spin moment $S_L(\Delta) = 0.65$, the same as the experimental one, can be obtained with energy window 0.1 eV and the value of the constant $U = 0.5$ eV. This constant is comparable with the constant $I = 0.92$ eV obtained by Janak (1977) in the calculation of the static susceptibility of paramagnetic iron with energy bands the same as in the present work. The value $I = 0.92$ eV was also obtained by Gunnarsson (1976, 1977) and Andersen *et al* (1977). However, in contrast to I the constant U corresponds to stability of the paramagnetic state (the Stoner criterion is unfulfilled: $U\nu(\varepsilon_F) = 0.79 < 1$). Moreover, the criterion of local magnetism (2.20) is also unfulfilled: the limit value U_c is 0.68 eV. It is important to note that the value $S_L(\Delta) = 0.65$ is obtained in the local single-site scheme, which does not take into account the spatial correlations of the spin density (the short-range magnetic order effects). Note

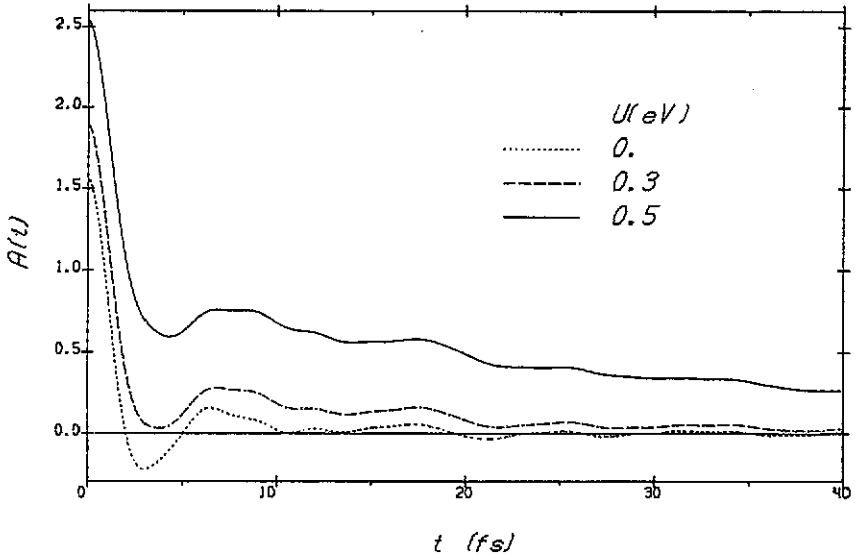


Figure 6. Time-dependent spin correlation function.

also that the magnitude of the local spin moment in the energy window 0.1 eV with constant $U = 0$ is only $S_L^0(\Delta) = 0.18$, which is smaller than the experimental value.

Finally, we take into consideration the time-dependent spin correlation function $A(t)$, which is obtained by inverse Fourier transformation of the function $A(\omega)$. Contributions to $A(t)$ are given by all harmonics of the correlation function $A(\omega)$, and therefore the form of the function $A(\omega)$ is essential for the time dependence $A(t)$. In figure 6 the calculated spin correlation function $A(t)$ is pictured for three values of the constant U : 0, 0.3 and 0.5 eV. The time correlation function for an electron system with constant $U = 0$ decreases very quickly from the value 1.57 at time $t = 0$ to zero at time $t_0 = 2$ fs. At larger times the function $A(t)$ has rapid oscillations and a damping tail. From the physical point of view, this means rather small damping time t_0 of the local spin moment created at $t = 0$. The damping time t_0 is comparable with the time τ_e of electron hopping from one site to another. This conformity is not accidental. Indeed, during the time τ_e an electron leaves a site and significantly changes the electron density.

Inclusion of electron-electron interaction results in slowing down of inter-site electron hopping processes and, eventually, in a considerable increase in the damping time of the local spin moment. For $U = 0.5$ eV the correlation function $A(t)$ decreases to zero in approximately $t_0 \approx 140$ fs. Thus, electron-electron interaction stabilizes the local spin moment, significantly slowing down the damping rate. For times considerably exceeding t_0 the usual oscillations and a damping tail are observed. Consider more closely the behaviour of the correlation function $A(t)$ at $t \leq t_0$. On the small time interval, ~ 4 fs, the correlation function decreases appreciably and then decreases very slowly. Such behaviour is connected with the contribution of two frequency regions of the correlation function $A(\omega)$. The sharp decrease at small times is determined by the total frequency region of the function $A(\omega)$, and the slope time is inversely proportional to the bandwidth. The region of slow decrease of the correlation function $A(t)$ is connected

with the resonance peak of $A(\omega)$ at low frequencies. Of course, it would not be quite correct to speak of the time t_0 as the damping time of the local spin moment. There is still the oscillating part, which also reflects the characteristic behaviour of the local spin moment. Moreover, any concrete time does not carry any specific sense in this context. Slow decrease of the correlation function $A(t)$ shows the importance of accounting for the time dispersion effects when developing more adequate theories of paramagnetic susceptibility.

5. Conclusions

In the present paper we have considered a simple model of accounting for electron correlation effects in calculations of the total single-site susceptibility and spin correlation function. If the usual RPA successfully describes spin excitations at low temperatures, when long-wave excitations are important, our single-site approximation (SSA) more exactly describes short-wave excitations at high temperatures, when any magnetic order is almost absent. The local dynamic susceptibility is connected with the single-site spin correlation function by the fluctuation-dissipation theorem, which allows one to calculate the latter. Results of calculations for paramagnetic iron have clearly shown the great importance of electron correlation effects in both local susceptibility and single-site spin correlation function. Electron correlation effects significantly change the frequency dependence of the local susceptibility, pumping over the susceptibility intensity from high to low frequencies. Similar behaviour is revealed for the local spin correlation function too. Electron-electron correlations, drastically changing the results of the one-electron approximation, allow one to describe on a quantitative level the experiments on polarized neutron scattering and to evaluate the damping time of local spin moments in the paramagnetic region ($t_0 \sim 10^{-13}$ s), which is two orders greater than the electron hopping time $\tau_e \sim 10^{-15}$ s. In this way, the static approximation of the spin fluctuation theories is substantiated in some degree. At the same time, when constructing the quantitative theory of different properties of transition metals, it will be necessary to account for the time variation of the electron spin density. Note also that the results of the present work were obtained without accounting for the short-range magnetic order.

Once again we stress the model character of the calculation of total local susceptibility, arising from the SSA with a single effective electron interaction constant U . However, qualitatively, and even quantitatively, the estimates obtained by us will scarcely change significantly, if susceptibility calculations are carried out on the full formula (2.10) with due regard for space and time fluctuations of electron spin density.

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