

## Magnetic susceptibility, exchange interactions and spin-wave spectra in the local spin density approximation

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2004 J. Phys.: Condens. Matter 16 7439

(<http://iopscience.iop.org/0953-8984/16/41/023>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.252.86.83

The article was downloaded on 27/05/2010 at 18:17

Please note that [terms and conditions apply](#).

# Magnetic susceptibility, exchange interactions and spin-wave spectra in the local spin density approximation

M I Katsnelson<sup>1,2</sup> and A I Lichtenstein<sup>2,3</sup>

<sup>1</sup> Department of Physics, Uppsala University, Box 530, SE-75121 Uppsala, Sweden

<sup>2</sup> Department of Physics, University of Nijmegen, NL-6525 ED Nijmegen, The Netherlands

<sup>3</sup> Institute of Theoretical Physics, University of Hamburg, Jungiusstrasse 9, 20355 Hamburg, Germany

Received 10 May 2004, in final form 15 July 2004

Published 1 October 2004

Online at [stacks.iop.org/JPhysCM/16/7439](http://stacks.iop.org/JPhysCM/16/7439)

doi:10.1088/0953-8984/16/41/023

## Abstract

Starting from an exact expression for the dynamical spin susceptibility in the time-dependent density functional theory, a controversial issue regarding exchange interaction parameters and spin-wave excitation spectra of itinerant electron ferromagnets is reconsidered. It is shown that the original expressions for exchange integrals based on the magnetic force theorem (Lichtenstein *et al* 1984 *J. Phys. F: Met. Phys.* **14** L125) are optimal for calculations of the magnon spectrum, whereas the static response function is better described using the ‘renormalized’ magnetic force theorem given by Bruno (2003 *Phys. Rev. Lett.* **90** 087205). This conclusion is confirmed by *ab initio* calculations for Fe and Ni.

An efficient scheme for making first-principles calculations of exchange interaction parameters in magnets based on a so-called ‘magnetic force theorem’ (MFT) [1, 2] in the density functional theory is frequently used for the analysis of exchange parameters for different classes of magnetic materials: dilute magnetic semiconductors [3], molecular magnets [4], colossal magnetoresistance perovskites [5], transition metal alloys [6], hard magnetic materials such as PtCo [7] and many others. Recently this method was generalized to take into account the correlation effects and successfully used for the quantitative estimation of exchange interactions in Fe and Ni [8, 9]. At the same time, the formal status of this approach is still not well defined, since a general mapping of formally rigorous spin density functional to an effective classical Heisenberg Hamiltonian can be made only approximately. It was noticed even in the first work on the MFT [1] that only the expression for spin-wave stiffness constant  $D$  is reliable. In terms of the diagrammatic many-body approach this means that the exchange integrals ( $J_{ij}$ ) should in general contain vertex corrections [10]. It is shown in [9] that the expressions of [1, 2] can be derived diagrammatically in a direct way from the calculations of the total energy

variation under spin rotations with the only assumption being that the vertex corrections can be neglected, without any reference to the magnetic force theorem. At the same time, using a general expression for spin-wave stiffness due to Hertz and Edwards [11], one can prove that the vertex corrections to  $D$  are cancelled for any local approximation for the self-energy (or, in the density functional method, for the local exchange–correlation potential) [12]. Recently Bruno has suggested [13] corrections to the MFT and consequently to the expressions for  $J_{ij}$  (see also [14, 15]). It is important to note that, first, the new expression for  $D$  coincides with the old one and, second, these corrections for the case of itinerant electron magnets are formally small in adiabatic parameter  $\eta = \varpi/\Delta$  where  $\varpi$  is a characteristic magnon frequency and  $\Delta$  is the Stoner spin splitting. At the same time, mapping of the local spin density approximation (LSDA) onto the classical Heisenberg model itself is valid only in the adiabatic approximation  $\eta \rightarrow 0$  [1, 2, 16]. If we are interested in higher-order effects in  $\eta$ , we might need different effective exchange parameters for different physical properties. We will show for few examples that this is exactly the case. It turns out that the spin-wave excitation spectrum should be calculated in terms of ‘old’ exchange integrals [1, 2], whereas for static properties the ‘new’ exchange integrals [13, 14] are more appropriate.

The most reliable way to consider spin-wave properties of itinerant electron magnets in the framework of the spin density functional theory is by the use of frequency-dependent magnetic susceptibility [17–19]. One should start from the time-dependent density functional theory in the adiabatic approximation (ADA-TDDFT) [20, 21]. We proceed with the Schrödinger-like equation within the self-consistent ADA-TDDFT potential

$$\begin{aligned} i\frac{\partial\psi}{\partial t} &= H\psi \\ H &= -\nabla^2 + V(\mathbf{r}) - \frac{1}{2}(\mathbf{B}_{\text{xc}}(\mathbf{r}) + \mathbf{B}_{\text{ext}}(\mathbf{r}))\sigma \end{aligned} \quad (1)$$

(Slater’s units are used here) where  $V(\mathbf{r})$  is an effective potential,  $\mathbf{B}_{\text{ext}}(\mathbf{r})$  and  $\mathbf{B}_{\text{xc}}(\mathbf{r})$  are external magnetic field acting on spin and exchange–correlation field, respectively. The adiabatic approximation means that the functional dependences of the exchange–correlation potential and field on the charge and spin density are supposed to be the same as in the stationary case. In the LSDA one has

$$\begin{aligned} V(\mathbf{r}) &= V_{\text{ext}}(\mathbf{r}) + \int d\mathbf{r}' \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \frac{\partial}{\partial n}[n\varepsilon_{\text{xc}}] \\ \mathbf{B}_{\text{xc}} &= -2\frac{\mathbf{m}}{m} \frac{\partial}{\partial m}[n\varepsilon_{\text{xc}}] \end{aligned} \quad (2)$$

where  $n$  and  $\mathbf{m}$  are charge and spin densities,  $\varepsilon_{\text{xc}}$  is the exchange–correlation energy per particle,  $V_{\text{ext}}$  is the external potential, i.e. the Coulomb potential of nuclei. To calculate the spin susceptibility we will assume  $\mathbf{B}_{\text{ext}}(\mathbf{r}) \rightarrow 0$ . This leads to the effective complete ‘non-equilibrium’ field

$$\delta B_{\text{tot}}^{\alpha} = \delta B_{\text{ext}}^{\alpha} + \frac{\delta B_{\text{xc}}^{\alpha}}{\delta m^{\beta}} \delta m^{\beta} \quad (3)$$

where  $\alpha\beta$  are Cartesian indices and the sum over repeated indices is assumed.

By definition of the exact non-local frequency-dependent spin susceptibility  $\hat{\chi}^{\alpha\beta}$ , the variation of the spin density is equal to

$$\delta m^{\alpha} = \hat{\chi}^{\alpha\beta} \delta B_{\text{ext}}^{\beta}. \quad (4)$$

The operator product is defined here as usual:

$$(\hat{\chi}\varphi)(\mathbf{r}) = \int d\mathbf{r}' \chi(\mathbf{r}, \mathbf{r}')\varphi(\mathbf{r}'). \quad (5)$$

On the other hand, it was shown in [20] that in the time-dependent density functional theory we should have exactly

$$\delta m^\alpha = \hat{\chi}_0^{\alpha\beta} \delta B_{\text{tot}}^\beta \quad (6)$$

where  $\hat{\chi}_0^{\alpha\beta}$  is the susceptibility of an auxiliary system of free Kohn–Sham particles. This was demonstrated originally for the charge excitations but the generalization of TDDFT for the spin-polarized case [21], shows that equation (6) holds also for spin excitations. Comparing the two expressions for  $\delta m^\alpha$  we have the following equation:

$$\hat{\chi}^{\alpha\beta} = \hat{\chi}_0^{\alpha\beta} + \hat{\chi}_0^{\alpha\gamma} \frac{\delta B_{\text{xc}}^\gamma}{\delta m^\delta} \hat{\chi}^{\delta\beta}. \quad (7)$$

This ‘RPA-like’ equation is formally exact in ADA-TDDFT. For the *local* spin density approximation (equation (2)) one has

$$\frac{\delta B_{\text{xc}}^\gamma}{\delta m^\delta} = \frac{B_{\text{xc}}}{m} \left( \delta_{\gamma\delta} - \frac{m^\gamma m^\delta}{m^2} \right) + \frac{m^\gamma m^\delta}{m^2} \frac{\partial B_{\text{xc}}}{\partial m}. \quad (8)$$

The first term in equation (8) is purely transverse and the second one is purely longitudinal with respect to the local magnetization vector. In collinear magnetic structures there are no couplings between the longitudinal ( $m^z$ ) and transverse ( $m^\pm = m^x \pm im^y$ ) components and for the transverse dynamical spin susceptibility dependent on the frequency  $\omega$  we have the following equation:

$$\chi^{+-}(\mathbf{r}, \mathbf{r}', \omega) = \chi_0^{+-}(\mathbf{r}, \mathbf{r}', \omega) + \int d\mathbf{r}'' \chi_0^{+-}(\mathbf{r}, \mathbf{r}'', \omega) I_{\text{xc}}(\mathbf{r}'') \chi^{+-}(\mathbf{r}'', \mathbf{r}', \omega) \quad (9)$$

where

$$I_{\text{xc}} = \frac{B_{\text{xc}}}{m} \quad (10)$$

is an exchange–correlation ‘Hund’s rule’ interaction, the magnetic and charge electron density being defined as usual:

$$\begin{aligned} m &= \sum_{\mu\sigma} \sigma f_{\mu\sigma} |\psi_{\mu\sigma}(\mathbf{r})|^2 \\ n &= \sum_{\mu\sigma} f_{\mu\sigma} |\psi_{\mu\sigma}(\mathbf{r})|^2. \end{aligned} \quad (11)$$

The bare susceptibility has the following form:

$$\chi_0^{+-}(\mathbf{r}, \mathbf{r}', \omega) = \sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \psi_{\mu\uparrow}^*(\mathbf{r}) \psi_{\nu\downarrow}(\mathbf{r}) \psi_{\nu\downarrow}^*(\mathbf{r}') \psi_{\mu\uparrow}(\mathbf{r}') \quad (12)$$

where  $\psi_{\mu\sigma}$  and  $\varepsilon_{\mu\sigma}$  are eigenstates and eigenvalues for the Kohn–Sham quasiparticles:

$$\begin{aligned} (H_0 - \frac{1}{2}\sigma B_{\text{xc}}) \psi_{\mu\sigma} &= \varepsilon_{\mu\sigma} \psi_{\mu\sigma} \\ H_0 &= -\nabla^2 + V(\mathbf{r}) \end{aligned} \quad (13)$$

and  $f_{\mu\sigma} = f(\varepsilon_{\mu\sigma})$  is the Fermi distribution function.

The derivation of the longitudinal spin susceptibility is similar to those presented above with a small complication, since we have to consider separately the responses of spin-up and spin-down electrons. Suppose we have an external perturbation  $\delta V_{\text{ext}}^\sigma$ . This leads to a change of the exchange–correlation potential  $V_{\text{xc}}^\sigma = \frac{\partial(n\varepsilon_{\text{xc}})}{\partial n_\sigma}$  ( $n_\sigma = \frac{1}{2}(n + \sigma m)$ ), namely,

$$\begin{aligned} \delta V_{\text{xc}}^\sigma &= U_{\sigma\sigma'} \delta n_{\sigma'} \\ U_{\sigma\sigma'} &= \frac{\partial^2(n\varepsilon_{\text{xc}})}{\partial n_\sigma \partial n_{\sigma'}} \end{aligned} \quad (14)$$

which gives the total perturbation  $\delta V_{\text{tot}}^\sigma = \delta V_{\text{ext}}^\sigma + \delta V_{\text{xc}}^\sigma$ . One can introduce the response functions  $K^{\sigma\sigma'}$  via the formal expression

$$\delta n^\sigma = K^{\sigma\sigma'} \delta V_{\text{ext}}^{\sigma'}. \quad (15)$$

Then the longitudinal spin susceptibility can be expressed in terms of the  $K$ -functions as

$$\chi^{zz} = \frac{1}{4} (K^{\uparrow\uparrow} + K^{\downarrow\downarrow} - K^{\uparrow\downarrow} - K^{\downarrow\uparrow}). \quad (16)$$

At the same time, according to the general scheme of TDDFT, we have

$$\delta n^\sigma = \hat{X}_\sigma \delta V_{\text{tot}}^\sigma \quad (17)$$

where

$$X_\sigma(\mathbf{r}, \mathbf{r}') = \sum_{\mu\nu} \frac{f_{\mu\sigma} - f_{\nu\sigma}}{\omega - \varepsilon_{\mu\sigma} + \varepsilon_{\nu\sigma}} \psi_{\mu\sigma}^*(\mathbf{r}) \psi_{\nu\sigma}(\mathbf{r}) \psi_{\mu\sigma}(\mathbf{r}') \psi_{\nu\sigma}^*(\mathbf{r}'). \quad (18)$$

Comparing these two expressions for  $\delta n^\sigma$  one obtains

$$\begin{aligned} K^{\uparrow\uparrow} &= X_\uparrow + X_\uparrow U_{\uparrow\uparrow} K^{\uparrow\uparrow} + X_\uparrow U_{\uparrow\downarrow} K^{\downarrow\uparrow} \\ K^{\downarrow\downarrow} &= X_\downarrow + X_\downarrow U_{\downarrow\downarrow} K^{\downarrow\downarrow} + X_\downarrow U_{\downarrow\uparrow} K^{\uparrow\downarrow} \\ K^{\uparrow\downarrow} &= X_\uparrow U_{\uparrow\downarrow} K^{\downarrow\downarrow} + X_\uparrow U_{\uparrow\uparrow} K^{\uparrow\downarrow} \\ K^{\downarrow\uparrow} &= X_\downarrow U_{\downarrow\uparrow} K^{\uparrow\uparrow} + X_\downarrow U_{\downarrow\downarrow} K^{\downarrow\uparrow}. \end{aligned} \quad (19)$$

Similar expressions have been obtained in the RPA for the Hubbard model in [22]. A coupling between the longitudinal spin and density degrees of freedom is important also for the electronic structure calculations which take into account correlation effects [9, 23].

Let us continue the derivation of useful expression for the transverse susceptibility (equation (9)). In order to consider the case of small  $\omega$  it is useful to make some identical transformations of the kernel (12), similar to the Hubbard model considerations [24]. Using equation (13) one can find

$$B_{\text{xc}} \psi_{\mu\uparrow} \psi_{\nu\downarrow}^* = (\varepsilon_{\nu\downarrow} - \varepsilon_{\mu\uparrow}) \psi_{\nu\downarrow}^* \psi_{\mu\uparrow} + \nabla(\psi_{\mu\uparrow} \nabla \psi_{\nu\downarrow}^* - \psi_{\nu\downarrow}^* \nabla \psi_{\mu\uparrow}). \quad (20)$$

Substituting equation (20) into (12) we obtain

$$(\chi_0^{+-} B_{\text{xc}})(\mathbf{r}, \mathbf{r}', \omega) = m(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}') - \omega \chi_0^{+-}(\mathbf{r}, \mathbf{r}', \omega) \quad (21)$$

where we used the completeness condition

$$\sum_{\mu} \psi_{\mu\sigma}^*(\mathbf{r}) \psi_{\mu\sigma}(\mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}'). \quad (22)$$

Substituting equation (21) into (12) we can transform the latter expression to the following form:

$$\hat{\chi}^{+-} = \hat{\chi}_0^{+-} + \hat{\chi}_0^{+-} \frac{B_{\text{xc}}}{m} \hat{\chi}^{+-} = \hat{\chi}_0^{+-} + \hat{\chi}^{+-} - \omega \hat{\chi}_0^{+-} \frac{1}{m} \hat{\chi}^{+-} + \frac{\hat{\Lambda}}{m} \hat{\chi}^{+-} \quad (23)$$

or, equivalently,

$$\hat{\chi}^{+-} = m[\omega - (\hat{\chi}_0^{+-})^{-1} \hat{\Lambda}]^{-1} \quad (24)$$

where

$$\Lambda(\mathbf{r}, \mathbf{r}', \omega) = \sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \psi_{\mu\uparrow}^*(\mathbf{r}) \psi_{\nu\downarrow}(\mathbf{r}) \nabla[\psi_{\mu\uparrow}(\mathbf{r}') \nabla \psi_{\nu\downarrow}^*(\mathbf{r}') - \psi_{\nu\downarrow}^*(\mathbf{r}') \nabla \psi_{\mu\uparrow}(\mathbf{r}')]. \quad (25)$$

Using equations (12), (24) one has finally

$$\hat{\chi}^{+-} = (m + \hat{\Lambda})(\omega - I_{\text{xc}} \hat{\Lambda})^{-1} \quad (26)$$

which is exactly equivalent to equation (9) but much more suitable for investigation of the magnon spectrum. Spin-wave excitations can be separated from the Stoner continuum (e.g., paramagnons) only in the adiabatic approximation, which means the replacement  $\Lambda(\mathbf{r}, \mathbf{r}', \omega)$  by  $\Lambda(\mathbf{r}, \mathbf{r}', 0)$  in equation (26). Otherwise, one should just find the poles of the total susceptibility, and the whole concept of ‘exchange interactions’ is not uniquely defined. Nevertheless, *formally* we can introduce the effective exchange interactions via the quantities

$$\Omega(\mathbf{r}, \mathbf{r}', \omega) = I_{xc} \Lambda(\mathbf{r}, \mathbf{r}', \omega). \quad (27)$$

Substituting equation (20) into (26) we get

$$\Lambda(\mathbf{r}, \mathbf{r}', \omega) = \sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \psi_{\mu\uparrow}^*(\mathbf{r}) \psi_{\nu\downarrow} [B_{xc}(\mathbf{r}') - \varepsilon_{\nu\downarrow} + \varepsilon_{\mu\uparrow}] \psi_{\nu\downarrow}(\mathbf{r}') \psi_{\mu\uparrow}(\mathbf{r}'). \quad (28)$$

Therefore

$$\begin{aligned} \Omega(\mathbf{r}, \mathbf{r}', \omega) &= \frac{4}{m(\mathbf{r})} J(\mathbf{r}, \mathbf{r}', \omega) + I_{xc}(\mathbf{r}) \sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \\ &\quad \times (\varepsilon_{\mu\uparrow} - \varepsilon_{\nu\downarrow}) \psi_{\mu\uparrow}^*(\mathbf{r}) \psi_{\nu\downarrow}(\mathbf{r}) \psi_{\nu\downarrow}^*(\mathbf{r}') \psi_{\mu\uparrow}(\mathbf{r}') \end{aligned} \quad (29)$$

where an expression for frequency-dependent exchange interactions has the following form:

$$J(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{4} \sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \psi_{\mu\uparrow}^*(\mathbf{r}) B_{xc}(\mathbf{r}) \psi_{\nu\downarrow}(\mathbf{r}) \psi_{\nu\downarrow}^*(\mathbf{r}') B_{xc}(\mathbf{r}') \psi_{\mu\uparrow}(\mathbf{r}'). \quad (30)$$

The latter coincides with the exchange integrals [1, 2, 25] if we neglect the  $\omega$ -dependence. Since  $B_{xc} \sim m$  we have  $J \sim m^2$  and the expression (30) vanishes in the non-magnetic case, as it should. Using the identity (22) one can show that

$$\Omega(\mathbf{r}, \mathbf{r}', 0) = \frac{4}{m(\mathbf{r})} J(\mathbf{r}, \mathbf{r}', 0) - B_{xc}(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}'). \quad (31)$$

Note that for  $\omega = 0$  we have exactly

$$-I_{xc} \hat{\Lambda} = \hat{\Omega}$$

and the static susceptibility  $\hat{\chi}^{+-}(0)$  can be represented in the form

$$\hat{\chi}^{+-}(0) = m(\hat{\Omega}^{-1} - B_{xc}^{-1}) \quad (32)$$

which is equivalent to the result of [13]

$$\hat{\hat{\Omega}} = \hat{\Omega}(1 - B_{xc}^{-1} \hat{\Omega})^{-1} \quad (33)$$

for the renormalized exchange interactions if one defines them in terms of inverse *static* susceptibility [14, 15].

As was stressed above, for a generic case of an itinerant electron magnet it is impossible to introduce the effective exchange integrals and one should work with the generalized spin susceptibility. Any definition of the exchange integrals assumes the adiabatic approximation somewhere. For the spin-wave spectrum which is determined by the pole of the transverse susceptibility it is natural to formulate an ‘exchange concept’: neglect of the  $\omega$ -dependence in  $\hat{\Omega}$ . Then, by virtue of equation (26) the magnon frequencies are just eigenstates of the operator  $\hat{\Omega}(0)$  which exactly corresponds to the expression from the ‘old’ MFT exchange interactions [1, 2]. Note that for the long-wavelength limit  $\mathbf{q} \rightarrow 0$  this result turns out to be exact, which proves the above statement about the stiffness constant  $D$ : in the framework of the local approximation it is rigorous. Corrections to  $D$  from a *non-locality* of the exchange–correlation potential have been estimated recently [26] for Fe and Ni; they turned out to be small.

At the same time, if we are interested in the computations of the thermodynamic properties such as the Curie temperature  $T_C$ , the renormalized exchange integrals can really give more accurate results. One can introduce for the itinerant electron magnets the magnon-like operators

$$b_{\mathbf{q}} = \frac{1}{\sqrt{m_0}} S_{\mathbf{q}}^- \quad b_{\mathbf{q}}^\dagger = \frac{1}{\sqrt{m_0}} S_{\mathbf{q}}^+ \quad (34)$$

where  $m_0 = 2\bar{S}$  is the ground-state magnetization and write for the temperature dependence of the magnetization the Bloch-like expression

$$m(T) = m_0 - \sum_{\mathbf{q}} \langle b_{\mathbf{q}}^\dagger b_{\mathbf{q}} \rangle = m_0 + \frac{1}{m_0} \sum_{\mathbf{q}} \int_{-\infty}^{\infty} d\omega \frac{\text{Im} \chi^{+-}(\mathbf{q}, \omega)}{\exp(\omega/T) - 1} \quad (35)$$

(a similar approximation for the Hubbard model has been proposed in [24]). If we use the classical spin approximation usually exploited for first-principles estimations of the Curie temperature, we should replace the Planck function in equation (35) by its classical limit  $T/\omega$  which immediately gives (taking into account the Kramers–Kronig relations) the following expression for the Curie temperature:

$$\frac{1}{T_C} = \frac{1}{m_0^2} \sum_{\mathbf{q}} \chi^{+-}(\mathbf{q}, \omega = 0) \quad (36)$$

which is identical to the expression from [13] in terms of the *renormalized* exchange interactions. Note, however, that the quantum character of the spin (which can be taken into account only beyond the LSDA) is probably very much essential for a proper description of high-temperature magnetism of transition metals [27], which makes the problem of improvement of *classical* estimations of  $T_C$  less important.

In order to test different approximations to the exchange interactions we calculated spin-wave spectrum for iron and nickel using the LMTO-TB method [28]. The orthogonal LMTO representation was used and the calculation scheme was the following.

The matrix of the Green function in the s, p, d basis set is equal to

$$G_{\sigma}(\mathbf{k}, \omega_n) = [i\omega_n + \mu - H_{\text{LDA}}^{\sigma}(\mathbf{k})]^{-1} \quad (37)$$

where  $\mu$  is the chemical potential,  $\omega_n$  are Matsubara frequencies and  $H_{\text{LDA}}$  is the orthogonal LSDA Hamiltonian. We use the following approximation for the matrix  $I_{\text{xc}}$ :

$$V_{\text{xc}} \equiv m I_{\text{xc}} = H_{\text{LDA}}^{\uparrow}(0) - H_{\text{LDA}}^{\downarrow}(0). \quad (38)$$

The matrix of the LDA susceptibility has been calculated using the fast Fourier transform technique with  $k = (\mathbf{k}, \omega_n)$ :

$$\chi_0^{+-}(q) = - \sum_k G_{\uparrow}(k) * G_{\downarrow}(k+q). \quad (39)$$

The Fourier transform of the ‘bare’ exchange interactions  $\hat{J}(0)$  (30) is defined as

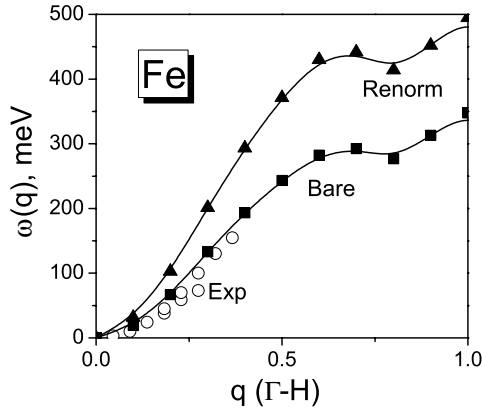
$$J(\mathbf{q}) = \frac{1}{4} V_{\text{xc}} \chi_0^{+-}(\mathbf{q}, \omega = 0) V_{\text{xc}} \quad (40)$$

whereas for the ‘renormalized’ exchange integrals [13, 14] one has

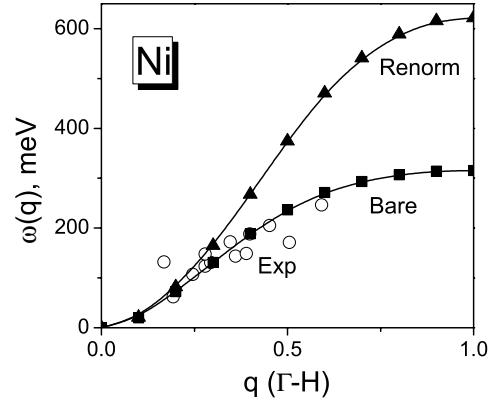
$$\begin{aligned} \tilde{J}(0) - \tilde{J}(\mathbf{q}) &= \frac{1}{4} \text{Tr}_L [m (\chi^{+-}(\mathbf{q}, \omega = 0))^{-1} m] \\ &= \frac{1}{4} \text{Tr}_L \left[ m \left( (\chi_0^{+-}(\mathbf{q}, \omega = 0))^{-1} - (\chi_0^{+-}(\mathbf{q} = 0, \omega = 0))^{-1} \right) m \right] \end{aligned} \quad (41)$$

where  $\text{Tr}_L$  means the trace over orbital indices. The magnon spectrum is determined via the exchange integrals as

$$\omega(\mathbf{q}) = \frac{4}{M} [J(0) - J(\mathbf{q})] \quad (42)$$



**Figure 1.** The spin-wave spectrum for ferromagnetic iron in the bare exchange (equation (40)) and renormalized exchange (equation (41)) schemes in comparison with experimental data (from [29]).



**Figure 2.** The spin-wave spectrum for ferromagnetic nickel in the bare exchange (equation (40)) and renormalized exchange (equation (41)) schemes in comparison with experimental data (from [29]).

**Table 1.** The Curie temperature (in K) for Fe and Ni calculated with the bare (equation (40)) and renormalized (equation (41)) LDA exchange interactions; ‘M’ is the mean-field approximation  $T_c = 2/3J(0)$  and ‘T’ is the Tjablikov, or RPA, approximation for  $T_c$ , similar to equation (36).

$T_c$ (K)	Exp.	Bare-M	Renorm.-M	Bare-T	Renorm.-T
Fe	1045	1060	1620	820	1280
Ni	631	310	760	285	630

where  $M = \text{Tr}_L(m)$  is the total magnetic moment. One can see from figures 1 and 2 that the LDA ‘bare’ exchange parameter describes the spin-wave spectra of Fe and Ni better, while the thermodynamics (e.g. the Curie temperature—see table 1) is more reasonable with the exact static LDA exchange (which is the ‘RPA’-like expression). Of course, one should not overestimate the accuracy of an approach based on adiabatic exchange parameters. As is emphasized above, there is no unique, unambiguous procedure for mapping the spin density functional to the classical Heisenberg model. It is especially important for discussion of calculated values of the Curie temperature. For example, the quantum character of the spin leads to enhancement of  $T_c$  by the factor  $(S+1)/S$  for the Heisenberg model with spin  $S$  in the mean-field approximation. A similar enhancement should be important also for itinerant electron magnets [1, 27]. The situation with the spin-wave spectrum is better, since the expression [1] for the stiffness constant appears to be exact in the local density approximation. This is also true in the dynamical mean-field theory for exchange interactions [8], as is proven in [12]. For finite magnon wavevectors, both ‘old’ [1, 2] and ‘new’ [13, 14] expressions should be considered as approximate ones in comparison with a more rigorous approach based on the calculations of dynamical transverse susceptibility. We have shown that the ‘new’ expression for exchange integrals does correspond better to the *static* magnetic susceptibility. However, the expression for the magnon frequency in terms of exchange integrals (42) is valid only in the Heisenberg model. For itinerant magnets, the ‘old’ exchange integrals for spin-wave spectra give better results due to an ‘error cancellation’, that is, cancellation of the renormalization of static susceptibility and of the residue of dynamic susceptibility in the magnon pole as is seen from equation (26). This conclusion is confirmed by our computational results for Fe and Ni.



## References

- [1] Lichtenstein A I, Katsnelson M I and Gubanov V A 1984 *J. Phys. F: Met. Phys.* **14** L125  
Lichtenstein A I, Katsnelson M I and Gubanov V A 1985 *Solid State Commun.* **54** 327
- [2] Lichtenstein A I, Katsnelson M I, Antropov V P and Gubanov V A 1987 *J. Magn. Magn. Mater.* **67** 65
- [3] Bouzerar G, Kudrnovsky J, Bergqvist L and Bruno P 2003 *Phys. Rev. B* **68** 081203
- [4] Boukhvalov D W *et al* 2002 *Phys. Rev. B* **65** 184435  
Boukhvalov D W *et al* 2004 *Phys. Rev. B* **70** 054417
- [5] Solovyev I V and Terakura K 2001 *Phys. Rev. B* **63** 174425  
Solovyev I V 2002 *Phys. Rev. B* **65** 144446
- [6] Umetsu R Y, Fukamichi K, Fujinaga Y and Sakuma A 2003 *J. Phys.: Condens. Matter* **15** 4589
- [7] Skomski R, Kashyap A and Sellmyer D J 2003 *IEEE Trans. Magn.* **39** 2917
- [8] Katsnelson M I and Lichtenstein A I 2000 *Phys. Rev. B* **61** 8906
- [9] Katsnelson M I and Lichtenstein A I 2002 *Eur. Phys. J. B* **30** 9
- [10] Aryasetiawan F and Karlsson K 1999 *Phys. Rev. B* **60** 7419
- [11] Hertz J A and Edwards D M 1973 *J. Phys. F: Met. Phys.* **3** 2174
- [12] Lichtenstein A I and Katsnelson M I 2002 *Band-Ferromagnetism. Ground-State and Finite-Temperature Phenomena (Springer Lecture Notes in Physics vol 580)* (Berlin: Springer) p 75
- [13] Bruno P 2003 *Phys. Rev. Lett.* **90** 087205
- [14] Antropov V P 2003 *J. Magn. Magn. Mater.* **262** L192
- [15] Szczech Y H, Tusch M A and Logan D E 1998 *J. Phys.: Condens. Matter* **10** 639
- [16] Antropov V P, Katsnelson M I, van Schilfgaarde M, Harmon B N and Kusnezov D 1996 *Phys. Rev. B* **54** 1019
- [17] Callaway J, Wang C S and Laurent D G 1981 *Phys. Rev. B* **24** 6491
- [18] Cooke J F, Lynn J W and Davis H L 1980 *Phys. Rev. B* **21** 4118  
Cooke J F, Blackman J A and Morgan T 1985 *Phys. Rev. Lett.* **54** 718
- [19] Savrasov S Y 1998 *Phys. Rev. Lett.* **81** 2570
- [20] Runge E and Gross E K U 1984 *Phys. Rev. Lett.* **52** 997  
Gross E K U and Kohn W 1990 *Advances in Quantum Chemistry* vol 21 (New York: Academic) p 255
- [21] Liu K L and Vosko S H 1989 *Can. J. Phys.* **67** 1015
- [22] Izuyama T, Kim D and Kubo R 1963 *J. Phys. Soc. Japan* **18** 1025
- [23] Katsnelson M I and Lichtenstein A I 1999 *J. Phys.: Condens. Matter* **11** 1037
- [24] Irkhin V Yu and Katsnelson M I 1990 *J. Phys.: Condens. Matter* **2** 7151
- [25] Lichtenstein A I, Anisimov V I and Zaanen J 1995 *Phys. Rev. B* **52** R5467
- [26] Katsnelson M I and Antropov V P 2003 *Phys. Rev. B* **67** R140406
- [27] Lichtenstein A I, Katsnelson M I and Kotliar G 2001 *Phys. Rev. Lett.* **87** 067205
- [28] Andersen O K and Jepsen O 1984 *Phys. Rev. Lett.* **53** 2571
- [29] Kübler J 2000 *Theory of Itinerant Electron Magnetism* (Oxford: Clarendon)