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Spin-wave excitation spectra of nickel and iron

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A Green's function formalism for calculating spin-wave excitations is developed for practical calculations and tested for real solids. The mapping to the Heisenberg Hamiltonian commonly used in spin-wave calculations is avoided, making the formalism suitable for both localized and itinerant magnetic systems. To test the formalism, we have calculated the spin-wave spectra and dispersions of ferromagnetic Fe and Ni. The results prove to be in very good agreement with experiment and some novel features are predicted.

First-principles studies of the electronic structure of materials have been primarily concentrated on the charge degree of freedom whereas the spin degree of freedom is principally frozen. The energy scale of spin fluctuations (a few tens of meV) is indeed much smaller than a typical energy scale of charge excitations which is of the order of a few eV. However, transport and thermal phenomena such as specific heat and resistivity are well-known examples which lie in the low-energy regime.[1] More recently, the study of spin excitations has gained a lot of interest due to the discovery of high-temperature superconductivity. There is compelling evidence that spin fluctuations are the mediator of the attractive interaction.[2,3]

Despite the importance of spin excitations, first-principles calculations using realistic energy bands and wave functions are rather rare. This is partly due to a lack of theoretical frameworks and partly due to a lack of efficient numerical schemes and the large numerical effort required. The most common technique for calculating spin-wave dispersions is the frozen magnon method where the problem is mapped to the celebrated Heisenberg Hamiltonian[4] and the parameters in the model are obtained from realistic calculations. Several shortcomings are readily apparent: The method does not give the spin-wave spectra so that lifetime and multiple branches are not accessible. Moreover, applications of the method to itinerant electrons are difficult to justify. Spin-wave excitation spectra are also attainable using time-dependent density functional theory (TD DFT).[5] Although the method is formally exact, the calculated spectra may depend significantly on the quality of the exchange-correlation potential. Thus, a simple approximation may not be adequate, and not surprisingly, the spin-wave dispersions calculated within the local density approximation[6–9] (LDA) are often in large discrepancy with experiment. A new method for calculating spin-wave spectra was also proposed recently but the applicability of the method has not been demonstrated.[10]

In this report, we develop a recently proposed formalism for calculating spin-wave excitations into a practical scheme and demonstrate its applicability by calculating the spin-wave spectra and dispersions of ferromagnetic Fe and Ni. The spin-wave spectra of these materials are rather complex and they should provide a stringent test of the method.

The experimental spin-wave dispersion of Ni (Ref. 11) shows a number of interesting features which have not been fully explained. A prominent feature is the presence of two branches, known as the ''acoustic'' and ''optical'' branches. Early pioneering calculations[12–14] and more recent studies[5] did succeed in reproducing the two branches but there are some problems. A more recent and probably the most up-to-date calculation using TD DFT gives a dispersion in significant deviation from experiment for the optical branch.[5] A possible explanation is that the Kohn-Sham band structure is in disagreement with the photoemission (quasiparticle) band structure. In particular, the exchange splitting, which is relevant for determining the spin-wave dispersion, is overestimated by the LDA by a factor of 2,[15,16] whereas this is not the case in Fe.

For Fe, existing calculations[5,12,17] have been successful in reproducing the experimental dispersion,[18,19] at least for small momentum transfer. However, a very interesting analysis of the temperature-dependent magnetization of Fe predicts a gap in the spin-wave dispersion at about 0.74 Å^{-1} which is necessary to explain the experimental magnetization data.[20] Such a crossover gap between the acoustic and optical branches is in fact not observed experimentally. As will be shown later, our calculations also predict a similar gap at the same wave vector.

We first describe the formalism and the numerical scheme. For details of the theory we refer to Ref. 21. Spin-wave excitations are related to the imaginary part of the magnetic τ-ordered response function[21]

$$\mathcal{R}_{ij}(\mathbf{r}\tau,\mathbf{r}'\tau') \equiv \frac{\delta\langle\hat{\sigma}^i(\mathbf{r}\tau)\rangle}{\delta\varphi_j(\mathbf{r}'\tau')}, \tag{1}$$

where $i,j=0,x,y,z$. The external field φ_j corresponds to a scalar one for $j=0$ and for $j=x,y,z$ to the magnetic field. Here α and β denote the spin and the spin density is given by

$$\langle\hat{\sigma}^i(\mathbf{r}\tau)\rangle = \sigma^i_{\beta\alpha}\mathcal{G}_{\alpha\beta}(\mathbf{r}\tau,\mathbf{r}\tau^+), \tag{2}$$

where a repeated index or variable implies a summation or integration provided the index or variable does not appear on the other side of the equation. $\mathcal{G}_{\alpha\beta}$ is the Matsubara (temperature) Green function and σ^i the Pauli spin matrices. From Ref. 21, the simplest approximation for $\mathcal{R}$ leads to

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$$\mathcal{R}_{ij}(\mathbf{q},\omega)=-\sigma^{i}_{\beta\alpha}\sigma^{j}_{\alpha\beta}\langle\mathbf{q}|B_{\mathbf{q}r}\rangle[\mathcal{K}^{\alpha\beta}_{rs}(\mathbf{q},\omega)+\mathcal{T}^{\alpha\beta}_{rs}(\mathbf{q},\omega)]\times\langle B_{\mathbf{q}s}|\mathbf{q}\rangle, \tag{3}$$

where $\langle\mathbf{q}|B_{\mathbf{q}r}\rangle=\int d^3r\exp(-i\mathbf{q}\cdot\mathbf{r})B_{\mathbf{q}r}(\mathbf{r})$, and schematically

$$\mathcal{T}=\mathcal{K}[1-\mathcal{W}K]^{-1}\mathcal{W}\mathcal{K}. \tag{4}$$

This is equivalent to the random phase approximation (RPA).[22] In our formalism, the screened interaction $\mathcal{W}$ emerges in a natural way as part of the theory. This is in contrast to other theories where the interaction between the electron is assumed to be screened. The interpretation of Eq. (3) is that $\mathcal{K}$ represents the noninteracting response and $\mathcal{T}$ describes correlations in the form of repeated particle-hole scattering events of opposite spins, which eventually is responsible for the formation of spin waves. The main task is to calculate the kernel $\mathcal{K}$ which is given by

$$\mathcal{K}^{\alpha\beta}(\mathbf{r},\mathbf{r}';\omega)=\sum_{\mathbf{k}\mathbf{k}'}\sum_{n}^{\mathrm{occ}}\sum_{n'}^{\mathrm{unocc}}\frac{\psi_{\mathbf{k}n\beta}(\mathbf{r})\psi^{*}_{\mathbf{k}'n'\alpha}(\mathbf{r})\psi_{\mathbf{k}'n'\alpha}(\mathbf{r}')\psi^{*}_{\mathbf{k}n\beta}(\mathbf{r}')}{(\omega+i\delta-\epsilon_{\mathbf{k}'n'\alpha}+\epsilon_{\mathbf{k}n\beta})}-\frac{\psi_{\mathbf{k}n\alpha}(\mathbf{r})\psi^{*}_{\mathbf{k}'n'\beta}(\mathbf{r})\psi_{\mathbf{k}'n'\beta}(\mathbf{r}')\psi^{*}_{\mathbf{k}n\alpha}(\mathbf{r}')}{(\omega+i\delta+\epsilon_{\mathbf{k}'n'\beta}-\epsilon_{\mathbf{k}n\alpha})}. \tag{5}$$

Here $\{\psi_{\mathbf{k}n\alpha},\epsilon_{\mathbf{k}n\alpha}\}$ are the LDA Bloch states and the corresponding eigenvalues, respectively. For practical calculations, it is suitable to use Bloch basis functions $B_{\mathbf{q}r}$ for the space variables. We have developed a scheme using a localized product basis,[23] based on the linear muffin-tin orbital (LMTO) method within the atomic sphere approximation.[24] The number of $\mathbf{k}$ points is ~ 1000 in the irreducible zone. The δ function in the imaginary part of kernel is replaced by a Gaussian $(\sqrt{\pi}\sigma)^{-1}\exp(-\omega^2/\sigma^2)$. We have used this technique rather than the tetrahedron method because we wish to investigate the effects of broadening on the spectra. In order to separate the band structure effects, we will solve for the response function using a *local* and *static* screened interaction $\mathcal{W}=\mathcal{W}(\mathbf{r},\mathbf{r}';\omega=0)\delta(\mathbf{r}-\mathbf{r}')$. The spin-wave spectra are given by $\mathrm{Im}\mathcal{R}_{-+}(\mathbf{q},\omega)\sim\mathrm{Im}\mathcal{R}_{xx}(\mathbf{q},\omega)$ and the spin-wave excitation energy $\omega_{\mathbf{q}}$ can be identified with the position of the main peak in $\mathrm{Im}\mathcal{R}_{-+}(\mathbf{q},\omega)$.

The experimental dispersion curve for Ni(100) is displayed in Fig. 1. The most recent calculation[5] using TD DFT is capable of reproducing the acoustic branch quite well but the calculated optical branch is in large discrepancy with experiment, as can be seen in Fig. 1. In an ordinary LDA calculation of Ni the exchange splitting for states at the top of the occupied band is overestimated by a factor of 2 compared to experiment (0.3 eV vs 0.6 eV).[15,16] To investigate the role of the one-particle band structure we first calculate the spin-wave spectra using the ordinary LDA band structure. The results are similar to those of Savrasov[5,25] as expected. In agreement with experiment,[11] the spin-wave dispersion curve exhibits two branches with the optical mode substantially too high. We now decrease the exchange splitting by one-half which has the consequence of reducing the energy for a Stoner spin-flip process also by a factor of one-half. The corresponding spin-wave dispersion curve is shown in Fig. 1. The existence of two spin-wave branches is still maintained but a drastic reduction in the energy of the optical spin-wave branch is found, leading to very good agreement with the experiment of Mook *et al.*[11]

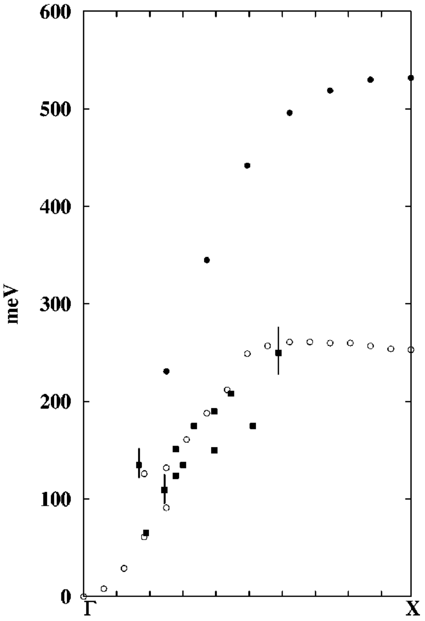

FIG. 1. Spin-wave dispersion along (100) for nickel. Open circles correspond to the present calculations and solid squares to experiment (Ref. 11). Experimental error bars are indicated by vertical solid lines. Also shown (black circles) is the optical mode obtained by Savrasov (Refs. 5 and 25).

The main difference between the calculated dispersion and the experiment is that the low-energy acoustic branch is disappearing for slightly smaller $\mathbf{q}$ values. Interesting to note is that the spin-wave excitation energy $\omega_{\mathbf{q}}$ is merely constant for $\mathbf{q}>0.6(1,0,0)2\pi/a$. The present results strongly indicate that the main source of problem in the TD DFT calculation can be traced back to defective LDA eigenvalues in Ni, in particular the exchange splitting. This is also supported by the fact that for Fe, where the LDA eigenvalues are in agreement with photoemission data, no reduction in the exchange splitting is necessary as shown later. Moreover, the TD DFT results for Fe are in good agreement with experiment.[5]

We now discuss the origin of the two branches. From Eq. (4) we notice that a double-peak structure in the spin-wave spectra may arise either from a complicated band structure implicitly contained in the kernel $\mathcal{K}$ or from a strong energy dependence in $\mathcal{W}$. Our results suggest that it is mainly the band structure that is responsible for the presence of the two branches since we have used a static $\mathcal{W}$. Typically, $\mathrm{Im}\mathcal{K}$ is peaked at about the average exchange-splitting energy corresponding to the Stoner spin-flip excitation where an electron from the majority channel is excited into the minority channel. Due to correlations, $(1-\mathcal{W}\mathrm{Re}\mathcal{K})$ and $\mathrm{Im}\mathcal{K}$ can both become zero or small at an energy below the Stoner peak. This gives rise to a collective excitation or spin waves. Generally speaking, the appearance of more than one spin-wave excitation, like the double-peak structure in the spin-wave spectra of Ni, implies the presence of additional structure in $\mathrm{Im}\mathcal{K}$ below the Stoner peak. In Fig. 2 we show a typical matrix element of $(1-\mathcal{W}\mathrm{Re}\mathcal{K})$ and $\mathrm{Im}\mathcal{K}$ for the case when we used a band structure with a reduced exchange splitting. Not dis-

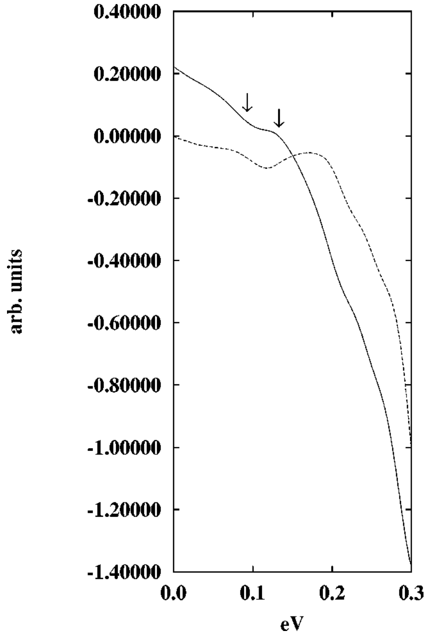

FIG. 2. Matrix element $i=j=1$ of $1-\mathcal{W}\mathrm{Re}\mathcal{K}$ (solid line) and $\mathcal{W}\mathrm{Im}\mathcal{K}$ (dashed line) for nickel; $\mathbf{q}=0.25(1,0,0)2\pi/a$. The vertical arrows show the peak positions in the corresponding spectra.

played is the Stoner peak located at approximately 0.3 eV. Additional structures below the Stoner peak are clearly visible which give rise to the double-peak structure leading to the two branches in the dispersion.

Due to a large density of states around the Fermi level which is mainly of 3d character, Stoner-flip d-d transitions are enhanced and result in the small peak around 0.1 eV in $\mathcal{W}\mathrm{Im}\mathcal{K}$. This in turn gives rise to a weak dip structure in $(1-\mathcal{W}\mathrm{Re}\mathcal{K})$ via the Kramers-Kronig relation. For $\mathbf{q}\sim(0.15\text{–}0.3,0,0)2\pi/a$ the dip structure in $(1-\mathcal{W}\mathrm{Re}\mathcal{K})$ is located close to zero and consequently there will be *two peaks* in the spin-wave excitation spectra. The presence of the double-peak structure is rather sensitive to both the position and intensity of the smaller peak. Thus for smaller and larger $\mathbf{q}$ values the dip structure does not give rise to any poles in $(1-\mathcal{W}\mathrm{Re}\mathcal{K})^{-1}$. For large $\mathbf{q}$, it is actually the position of the main peak in $\mathcal{W}\mathrm{Im}\mathcal{K}$ that determines the spin-wave spectra.

Considering the cubic symmetry of Ni, one expects the spin-wave dispersion to be isotropic. However, there is one puzzling feature in the spin-wave dispersion along the (111) direction: Only one branch is observed, and calculations by Savrasov[5] seem to confirm this. It is feasible that the double branch in the spin-wave dispersion is due to a complicated nesting structure of the Fermi surface which is sensitive to the direction in $\mathbf{k}$ space. On the other hand, the experimental resolution is better along (100) than (111), so that a narrow double-peak structure may not be observable.[11] Adopting the same reduction in the exchange splitting as in the (100) direction, we have calculated the spin-wave dispersion along (111) using several different broadenings. With $\sigma=0.08$ eV the spin-wave dispersion is in very good agreement with the experimental data.[11] However, using the same broadening as for the (100) direction ($\sigma=0.02$ eV) we can clearly distinguish a prominent double-peak in the spin-wave spectra as shown in Fig. 3. The results reveal that there is clearly an underlying double-peak structure which becomes smeared out when a larger broadening is used. Therefore we have strong reasons to believe that there also exist two branches along the (111) direction.

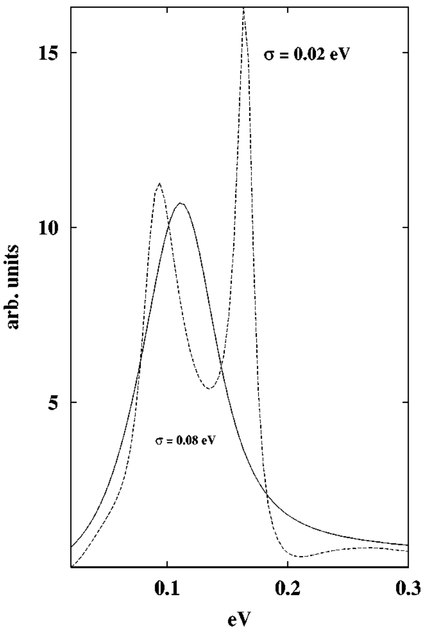

FIG. 3. Spin-wave excitation spectra of nickel for $\mathbf{q}=0.1875(1,1,1)2\pi/a$ for different broadenings.

The experimental spin-wave dispersion of Fe does not appear to show anything peculiar.[18,19] Unlike in Ni, the dispersion only shows one branch. The LDA exchange splitting for Fe compares well with the experimental one (2.2 eV), so no reduction for the exchange splitting is introduced. Up to the available experimental data taken by Lynn,[18] our results shown in Fig. 4 are in very good agreement with experiment. In fact, recent measurements[19] followed the spin-wave exci-

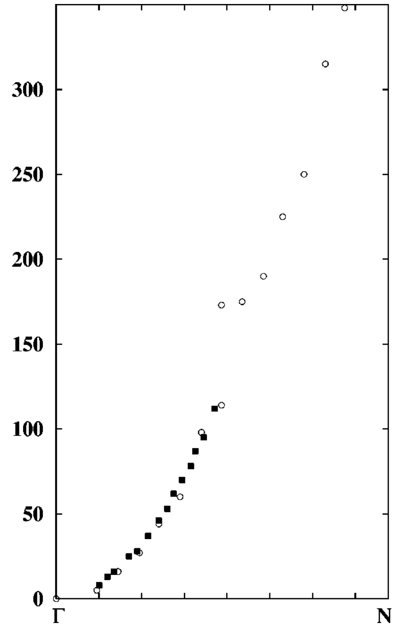

FIG. 4. Spin-wave dispersion along (110) for iron. Solid squares correspond to experiment (Ref. 18) and open circles to the present calculations.

tations to the zone boundary, and the observed energy was ~ 350 meV, in agreement with our prediction.

Within the experimental sensitivity there is no sign of two branches. But surprisingly, we found a gap in the spin-wave dispersion at $\mathbf{q}=0.25(1,1,0)2\pi/a$ with a substantially lower energy than the optical mode found in Refs. 12 and 17, as can be seen in Fig. 4. A very interesting work on the temperature-dependent magnetization of Fe and Ni was recently published by Ododo and Anyakoha[20] where they analyzed experimental data regarding the variation of magnetization with temperature. In order to explain the experimental magnetization curve, they concluded that there should exist a gap in the spin-wave dispersion of Fe at $\mathbf{q}=0.25(1,1,0)2\pi/a$ and 170 meV, in perfect agreement with our findings. Considering that the double-peak structure in the calculated spectra is not as prominent as in the case of Ni, it is not surprising that such a structure may be difficult to observe experimentally. Our calculations and the analysis of Ododo and Anyakoha,[20] however, give strong support for the presence of the gap in the dispersion.

In summary, we have developed a proposed formalism for calculating spin-wave spectra into a practical scheme and shown its practicability by calculating the spin-wave spectra and dispersions of Fe and Ni. With this scheme, the result for the optical branch of Ni is in significantly better agreement compared with previous calculations and additional features are uncovered which have not observed before either theoretically or experimentally.

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