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Electron-magnon interactions in Ni

Leonard Kleinman
Department of Physics, The University of Texas, Austin, Texas 78712
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We demonstrate that the minority-spin-electron (or majority-spin-hole) self-energy due to magnon exchange for the case of strong ferromagnetism can be calculated from the one-electron energy bands of the local-spin-density-functional Hamiltonian. This is a good model for Ni where we estimate both the real and imaginary parts of the majority-spin-hole self-energy to be less than 0.01 eV, thus demonstrating that many-body effects play no role in the photoelectron-spin-polarization reversal observed in Ni.

We have recently shown[1] that the photoelectron-spin-polarization reversal observed[2] 0.1 eV above threshold in Ni could be entirely explained by one-electron *surface*-energy-band theory. It was pointed out[3] to us that many-body effects were believed to be large and therefore the agreement between one-electron theory and experiment might be fortuitous. We argued that there were no propagating final states near threshold for (100) Ni and that the final state had to be an evanescent LEED state so that even if many-body effects were important, they had to be calculated at the surface. We further argued that strong many-body effects would smear out structure in the density of states (DOS) and that it appeared impossible to explain the extremely rapid rise in photoelectron spin polarization from negative to positive values (as a function of photon frequency) without having a very sharp peak in the majority-spin surface DOS, just below the Fermi energy. The question of the importance of many-body effects remained unresolved.

The many-body theories[4,5] are generally based upon the single-band Hubbard Hamiltonian which we believe to be an extremely inappropriate model for Ni for the following reason. The $^3F(3d^84s^2)$ and $^3D(3d^94s^1)$ configurations of an atomic Ni are essentially degenerate.[6] The single-band Hubbard model, in which the electrons interact via a strong short-range replusion when they occupy the same atomic orbital, ignores the possibility of a d electron being promoted to an s state at almost no cost in energy, undergoing an interaction which otherwise would be inhibited by the strong short-range repulsion seen by d electrons, and then returning to the d state. Because the d electrons in Ni are itinerant and because energy-band calculations[7] made within the spin-density-functional approximation[8] appear to be fairly accurate, we here base our calculation of the electron self-energy (due to magnon scattering) upon the local-spin-density-functional Hamiltonian[9]

$$H = -\tfrac{1}{2}\nabla^2 + V_0(\vec{r}) + V_f(\vec{r})\vec{\sigma}\cdot\hat{n}(\vec{r}) , \qquad (1)$$

where $V_0(\vec{r})$ contains the Coulomb and spin-averaged exchange and correlation potentials obtained from the ions and self-consistently calculated electron charge distribution and $V_f(\vec{r})$ represents the difference between the self-consistent up- and down-spin electron exchange and correlation potentials. In principle V_0 and V_f contain electron-magnon self-energy contributions but in practice of course, they do not. We shall here assume that V_0 and V_f include all exchange and correlation contributions except for the electron-magnon interaction and use Eq. (1) to determine the effects of that interaction. The unit vector $\hat{n}$ which normally points in the z direction may be written

$$\hat{n} = [1 - \tfrac{1}{2}(n_x^2 + n_y^2)]\hat{i}_z + n_x\hat{i}_x + n_y\hat{i}_y \qquad (2)$$

to second order in n_x and n_y. Defining

$$\sigma_\pm = \tfrac{1}{2}(\sigma_x \pm i\sigma_y) , \qquad (3)$$

$$\hat{n}_\pm = n_x \pm i n_y , \qquad (4)$$

we find the perturbing potential due to the deviation of $\hat{n}$ from the z direction is

$$V'(\vec{r}) = V_f(\vec{r})(-\tfrac{1}{2}\hat{n}_+\hat{n}_-\sigma_z + \hat{n}_+\sigma_- + \hat{n}_-\sigma_+) . \qquad (5)$$

We now assume the magnons are independent elementary excitations of the interacting electron system just as Bohm and Pines[10] assumed for plasmons. Then

$$\hat{n}_+ = 2N^{-1/2}\sum_{\vec{q}} a_{\vec{q}}\, e^{i\vec{q}\cdot\vec{r}} , \qquad (6a)$$

$$\hat{n}_- = 2N^{-1/2}\sum_{\vec{q}} a_{\vec{q}}^\dagger\, e^{-i\vec{q}\cdot\vec{r}} \qquad (6b)$$

where $a_{\vec{q}}$ and $a_{\vec{q}}^\dagger$ are magnon destruction and creation operators. Here N is the number of (majority spin[11]) electrons and the factor $2N^{-1/2}$ in (6a) and (6b) is required so that the creation of the magnon will reduce the z component of spin by unity.[12] Second quantizing the electronic part of $V'(\vec{r})$ and adding a magnon kinetic-energy term[13] we obtain

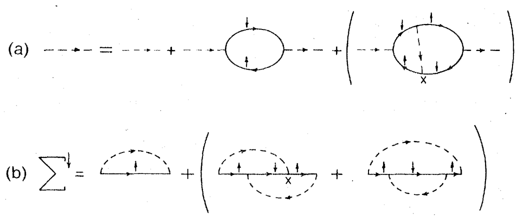

FIG. 1. (a) Magnon Dyson equation. The diagram in parenthesis does not exist because it fails to conserve spin at the vertex marked **×**. (b) The self-energy of an electron in the otherwise empty minority spin bands. The first diagram in parenthesis does not exist because it fails to conserve spin at the vertex marked **×**. The second diagram does not exist in the case of strong ferromagnetism studied here but does exist when magnons of spin+1 are present.

$$H' = \sum_{nn'\vec{k}\vec{k}'\vec{q}} (C^\dagger_{n\vec{k}\uparrow} C_{n'\vec{k}'\downarrow} a^\dagger_{\vec{q}} M_{n\vec{k}\uparrow,n'\vec{k}'\downarrow} \delta_{\vec{k}',\vec{k}+\vec{q}} + \text{H.c.}) - N^{1/2} \sum_{n\vec{k}\vec{q}} C^\dagger_{n\vec{k}\uparrow} C_{n\vec{k}\uparrow} a^\dagger_{\vec{q}} a_{\vec{q}} M_{n\vec{k}\uparrow,n\vec{k}\uparrow} + \sum_{q} \Omega_{\vec{q}} a^\dagger_{\vec{q}} a_{\vec{q}} , \qquad (7)$$

where $C^\dagger_{n\vec{k}\sigma}$ creates an electron (or destroys a hole) in the nth band with z component of spin $\sigma = +\frac{1}{2}$ or $-\frac{1}{2}$ and wave vector $\vec{k}$ lying within the first Brillouin zone.[14] The matrix element is given by

$$M_{n\vec{k}\sigma,n'\vec{k}'\sigma'} = 2N^{-1/2} \int V_f(\vec{r}) u^*_{n\vec{k}\sigma}(\vec{r}) u_{n'\vec{k}'\sigma'}(\vec{r})\, d^3r , \qquad (8)$$

where $u_{n\vec{k}\sigma}(\vec{r})$ is the periodic part of the $(n\vec{k}\sigma)$ Bloch function. Noting that $M_{n\vec{k}\sigma,n\vec{k}\sigma} = -N^{-1/2}\Delta(n\vec{k})$ where $\Delta(n\vec{k})$ is the exchange splitting, we see that the last two terms in H' are just

$$H'_{\text{magnon}} = (\Delta + \Omega_{\vec{q}}) a^\dagger_{\vec{q}} a_{\vec{q}} , \qquad (9)$$

where Δ is the average $\Delta(n, \vec{k})$.

The magnon Green's function is defined as

$$D(\vec{q}, t) = -i \langle 0 \,|\, T[a_{\vec{q}}(t) a^\dagger_{\vec{q}}(0)] \,|\, 0 \rangle , \qquad (10)$$

where T is the time ordering operator. This differs from the corresponding phonon Green's function because of the lack of the spin nonconserving term[15]

$$C^\dagger_{n'\vec{k}'\uparrow} C_{n\vec{k}\downarrow} a_{\vec{q}} M_{n'\vec{k}'\uparrow,n\vec{k}\downarrow} \delta_{\vec{k}',\vec{k}+\vec{q}}$$

and its Hermitian conjugate in Eq. (7). The time Fourier transform of (10) for noninteracting magnons is

$$D_0(\vec{q}, q_0) = (q_0 - \Delta - \Omega_{\vec{q}} + i\delta)^{-1} , \qquad (11)$$

where δ is a positive infinitesimal. The Dyson equation for the clothed magnon propagator

$$D(\vec{q}, q_0) = [q_0 - \Delta - \Omega_{\vec{q}} - \tau(\vec{q}, q_0) + i\delta]^{-1} \qquad (12)$$

is diagramatically displayed in Fig. 1(a). We have that

$$\tau(q, q_0) = -i \sum_{nn'} (2\pi)^{-4} \int d^4k \, | M_{n\vec{k}\uparrow,n'\vec{k}+\vec{q}\downarrow} |^2 \times G_0^\uparrow(n, k) G_0^\downarrow(n', k+q) , \qquad (13)$$

where the

$$G_0^\sigma(n, k) = (k_0 - \epsilon^\sigma_{n\vec{k}} + i\eta^\sigma_{n\vec{k}})^{-1} \qquad (14)$$

are the one-electron propagators for the eigenstates of Eq. (1) with energy $\epsilon^\sigma_{n\vec{k}}$ and $\eta^\sigma_{n\vec{k}}$ is a positive (negative) infinitesimal for $\epsilon^\sigma_{n\vec{k}} > E_F (< E_F)$. Note that $\eta^\downarrow_{n'\vec{k}+\vec{q}}$ is always positive because we have assumed[11] the minority spin bands lie above E_F. The k_0 integration in (13) thus yields a nonzero result only if $\eta^\uparrow_{nk}$ is negative and we obtain

$$\tau(\vec{q}, q_0) = (2\pi)^{-3} \sum_{nn'} \int d^3k \, | M_{n\vec{k}\uparrow,n'\vec{k}+\vec{q}\downarrow} |^2 (q_0 + \epsilon^\uparrow_{n\vec{k}} - \epsilon^\downarrow_{n'\vec{k}+\vec{q}} + i\delta)^{-1} f^\uparrow_n(\vec{k}) [1 - f^\downarrow_{n'}(\vec{k}+\vec{q})] , \qquad (15)$$

where the $f^\sigma_n(\vec{k})$ are zero above E_F and unity below.

Although $\tau(\vec{q}, q_0)$ could in principle be calculated exactly from the energy bands obtained from Eq. (1), we will limit ourselves to a free-electron approximation with

$$\epsilon^\uparrow_{n\vec{k}} = (2m^*)^{-1} k^2 - \tfrac{1}{2}\Delta ,$$

$$\epsilon^\downarrow_{n\vec{k}+\vec{q}} = (2m^*)^{-1} (\vec{k}+\vec{q})^2 + \tfrac{1}{2}\Delta ,$$

and $M_{n\vec{k}\sigma,n'\vec{k}'\sigma'} = -N^{-1/2}\Delta$. Thus, we obtain

$$\tau(\vec{q}, q_0) = \tfrac{3}{2} \frac{m^*\Delta^2}{k_F^2} \left\{ m^* \frac{q_0 - \Delta}{q^2} - \frac{1}{2} + \frac{k_F}{2q} \left[1 - \left(m^* \frac{q_0 - \Delta}{k_F q} - \frac{q}{2k_F} \right)^2 \right] \ln \frac{2m^*(q_0 - \Delta) - q^2 + 2k_F q + i\eta}{2m^*(q_0 - \Delta) - q^2 - 2k_F q + i\eta} \right\} \qquad (16)$$

where we have used $N = k_F^3/6\pi^2$. Because the clothed magnon frequency $\omega_{\vec{q}}$ may be obtained from the pole in the transverse magnetic susceptibility $\chi(q, \omega_{\vec{q}})$ [which also can be calculated[9] from the energy bands obtained from Eq. (1)], we see that $\Omega_{\vec{q}}$ is obtainable from those energy bands through the relation

$$\omega_{\vec{q}} = \Delta + \Omega_{\vec{q}} + \tau(\vec{q}, \omega_{\vec{q}}) . \tag{17}$$

In the free-electron limit the pole in $\chi(\vec{q}, \omega_{\vec{q}})$ occurs[16] when $\Delta + \tau(\vec{q}, \omega_{\vec{q}}) = 0$, so in that limit $\Omega_{\vec{q}} = \omega_{\vec{q}}$. We note that diagrams like the last one in Fig. 1(a) (magnon vertex corrections) do not contribute to τ because they cannot conserve spin at both vertices of the interior magnon.

Now that we have magnon propagator we may proceed to calculate the self energy of minority spin electron states shown diagramatically in Fig. 1(b). We have

$$\Sigma^{\downarrow}(n, \vec{p}, p_0) = \frac{i}{2\pi} \sum_{n'\vec{q}} |M_{n\vec{p}\downarrow, n'\vec{p}-\vec{q}\uparrow}|^2 \int_{-\infty+i\delta}^{\infty+i\delta} \frac{dq_0}{[q_0 - \Delta - \Omega_{\vec{q}} - \tau(\vec{q}, q_0)](p_0 - q_0 - \epsilon^{\uparrow}_{n',\vec{p}-\vec{q}} + i\eta^{\uparrow}_{n',\vec{p}-\vec{q}})}$$
$$= \sum_{n'\vec{q}} \frac{[1 - f^{\uparrow}_{n'}(\vec{p} - \vec{q})]\,|M_{n\vec{p}\downarrow, n'\vec{p}-\vec{q}\uparrow}|^2}{p_0 - \epsilon^{\uparrow}_{n',\vec{p}-\vec{q}} - \Omega_{\vec{q}} - \Delta - \tau(\vec{q}, p_0 - \epsilon^{\uparrow}_{n',\vec{p}-\vec{q}}) + i\eta} . \tag{18}$$

In obtaining (18) we had to close the path of integration in the positive imaginary half-plane because of the branch cut along the real axis introduced by τ. Note that the third diagram on the right side of Fig. 1(b), a vertex correction with a backward propagating magnon, is not allowed because it fails to conserve spin at the vertex marked with an x. The last diagram, which also contains a backward propagating magnon, is allowed but only when spin +1 magnons are present.[15]

If we assume we have a single band of magnons with infinite lifetimes (i.e., real τ), $\mathrm{Im}\Sigma^{\downarrow}(n, \vec{p}, p_0)$ can be estimated by replacing $M_{n\vec{p}\downarrow, n'\vec{p}-\vec{q}\uparrow}$ by its average value,[17] $M = -(5N)^{-1/2}\Delta$ and assuming the $\vec{q}$ dependence of $\Omega_{\vec{q}} + \tau(\vec{q}, \vec{p}_0 - \epsilon_{n',\vec{p}-\vec{q}})$ is negligible compared to that of $\epsilon^{\uparrow}_{n',\vec{p}-\vec{q}}$. We then have

$$\Sigma^{\downarrow}(p_0) = M^2 \int_{E_F}^{E^{\uparrow}_{\max}} \frac{(dN^{\uparrow}/d\epsilon)\, d\epsilon}{p_0 - \epsilon - \Delta - \Omega - \tau(p_0 - \epsilon) + i\eta} , \tag{19}$$

yielding

$$\mathrm{Im}\,\Sigma^{\downarrow}(p_0) = -\tfrac{1}{5}\, \pi\Delta^2 M^{-1} (dN^{\uparrow}/d\epsilon)_{p_0-\omega} \times [1 - (d\tau/dq_0)_{\omega}]^{-1} \Theta(p_0 - \omega - E_F) , \tag{20}$$

where ω is an average $\omega_{\vec{q}}$ and Θ is a smeared out step function.[18] We can estimate the renormalization factor $[1 - (d\tau/dq_0)]^{-1}$ by noting that because $\mathrm{Re}\tau + \Delta$ vanishes on the energy shell in the free-electron limit, $\mathrm{Re}\tau$ can be expanded in a Taylor series

$$\mathrm{Re}\tau(\vec{q}, q_0) = \sum_{n=1} a_n (q_0 - \omega_{\vec{q}})^n - \Delta . \tag{21}$$

An expansion of (16) in the small q_0 and $\vec{q}$ limit yields $\omega_{\vec{q}} = (1 - 2k_F^2/5\Delta)q^2$ and $a_1 = -1$. Thus $(d\tau/dq_0)_{\omega} = -1$ for any $\vec{q}$ in the free-electron limit and the renormalization factor is simply $\frac{1}{2}$. For the remaining factors in (20) we need not rely on the free-electron approximation. Inserting values appropriate[19] to Ni $dN^{\uparrow}/d\epsilon = 30$ electrons per atom per Hartree, $N = 0.56$ electrons per atom, and $\Delta = 0.5$ eV, we obtain for a typical value,

$$\mathrm{Im}\Sigma^{\downarrow} \approx 0.15 \text{ eV}. \tag{22}$$

$\mathrm{Re}\Sigma^{\downarrow}(p_0)$ may be obtained from the Kramers-Kronig relation.[20] It peaks where $\mathrm{Im}\Sigma^{\downarrow}$ varies most rapidly, i.e., for[18] $E_F < p_0 < E_F + \omega_{\max}$. If $\omega_{\max}$ is larger than 0.2 eV, then $|\mathrm{Re}\Sigma^{\downarrow}|$ will not exceed 0.2 eV. Due to finite magnon lifetimes the intensity of neutron scattering[21] by Ni magnons with wave vector $q = 0.27\ q_{Bz}$ where q_{Bz} is the zone-boundary wave vector[22] (in any of the three principal directions) is about 2% of that by small $\vec{q}$ magnons. Thus $\tau(\vec{q}, p_0 - \epsilon_{n',\vec{p}-\vec{q}})$ in Eq. (18) has a large imaginary part for $\vec{q}$ in the 98% of the zone with $q > 0.27\ q_{Bz}$. This will cause $\mathrm{Im}\Sigma^{\uparrow}(n, \vec{p}, p_0)$[19] to be considerably smaller than our estimate in Eq. (22). It will also smooth out the structure in $\mathrm{Im}\Sigma^{\uparrow}(n, \vec{p}, p_0)$ making an even larger reduction in $\mathrm{Re}\Sigma^{\uparrow}(n, \vec{p}, p_0)$. Thus we think it quite likely that both the real and imaginary parts of $\Sigma^{\uparrow}$ are less than 0.01 eV in nickel. (We of course refer only to the magnon contribution to $\Sigma^{\uparrow}$. The imaginary part of $\Sigma^{\uparrow}$ for low-lying holes in Ni is so large that the lower part of the photoelectron distribution curve is completely wiped out. This probably involves a process in which a $3d$ electron drops into the $3d$ hole and excites another $3d$ electron into the nearly-free-electron bands without any spin reversals. This contribution to $\Sigma^{\uparrow}$ should be very small near the top of the majority d band which is the region of interest for explaining the photoemission polarization reversal.)

Thus, we have demonstrated that the self-energy of minority spin electrons due to magnon exchange can be calculated directly from the energy bands arising from the one-electron spin density functional Hamiltonian, at least for the case of strong ferromagnetism where the bottom of the minority band lies above the Fermi energy. The same theory applies to the majority spin holes in Ni because the top of their band lies below E_F (ignoring the s and p majority spin states except for their possible effect on magnon lifetime). Our estimate of the Ni majority spin hole self energy, though crude, is sufficiently accurate to demonstrate that its contribution to

$$G^{\uparrow}(n,\vec{k},k_0)=[k_0-\epsilon^{\uparrow}_{n\vec{k}}-\Sigma^{\uparrow}(n,\vec{k},\vec{k}_0)+i\eta^{\uparrow}_{n\vec{k}}]^{-1} \quad (23)$$

and hence to the majority spin density of states

$$\rho^{\uparrow}(E)=-\frac{1}{\pi}\sum_{\vec{k}n}\mathrm{Im}G^{\uparrow}(n,\vec{k},E) \quad (24)$$

is negligible. Therefore the belief that $\Sigma^{\uparrow}(n,\vec{k},\vec{k}_0)$ causes the majority spin density of states to have a large value at or immediately below E_F is shown to be incorrect. The one-electron surface-state explanation of the photoelectron spin polarization reversal appears to be not only sufficient but also necessary.

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[6]The ground state of Ni is $^3F(3d^84s^2)$; however, the $^3D(3d^94s^1)$ configuration has lower energy when the three spin-orbit split levels of both configurations are averaged over.

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[10]D. Bohm and D. Pines, Phys. Rev. 92, 609 (1953). Note that the two conjugate field operators in our case are n_x and n_y, which obey the commutation relation $[n_x(\vec{r}),\, n_y(\vec{r}\,')]=2i\delta(\vec{r}-\vec{r}\,')$.

[11]We will assume we have only majority-spin electrons (or in the case of Ni minority-spin holes) in the ground state. If both majority- and minority-spin states exist at the Fermi energy, $\hat{n}$ in Eq. (1) must be replaced by $\vec{M}/M_0$ to allow for spin waves in the minority-spin band which increase the magnetization $\vec{M}$.

[12]From Eqs. (5), 6(a), 6(b) and $a_{\vec{q}}a^{\dagger}_{\vec{q}}=n_{\vec{q}}+1$, we have $\Delta n_z=-2N^{-1}(n_{\vec{q}}+1)$. Since the reversal of a single spin reduces N to $N-2$, we see that Δn_z goes in steps of $2/N$, as is required.

[13]The function $\Omega_{\vec{q}}$ will shortly be obtained. The $\Omega_{\vec{q}}$ term in Eq. (7) may be thought of as arising from a series of grad $\hat{n}$ terms added to Eq. (1), the first of which would be of the form $\int d^3r[(\nabla n_x)^2+(\nabla n_y)^2]$.

[14]The $\delta_{\vec{k}',\vec{k}+\vec{q}}$ implies $\vec{k}'=\vec{k}+\vec{q}$ mod reciprocal-lattice vector.

[15]Because of interactions between phonons traveling forward and backward in time it is convenient to define the phonon propagator as the sum of forward and backward propagators, $D=-i\langle 0|T[a_{\vec{q}}(t)a^{\dagger}_{\vec{q}}(0)+a^{\dagger}_{\vec{q}}(t)a_{\vec{q}}(0)]|0\rangle$. Because of the lack of the spin-nonconserving terms, backward traveling magnons do not exist. They would, however, exist if we had both minority and majority spin electrons at the Fermi energy, and hence had magnons with both spin+1 and −1. For an interesting discussion see R. D. Mattuck, *A Guide to Feynman Diagrams in the Many-Body Problem*, (McGraw-Hill, New York, 1967), Chap. 16.

[16]In the free-electron limit $\chi=\chi_0(1-\lambda\chi_0)^{-1}$. It may easily be seen from Ref. 9 that $-\lambda\chi_0=\tau/\Delta$.

[17]That the average matrix element is equal to the free-electron matrix element divided by the square root of the number of d bands of one spin can be seen as follows. Consider Eq. (8) for $\vec{k}=\vec{k}'=0$; then by symmetry $M_{n0\sigma,n'0\sigma'}=-N^{-1/2}\Delta(n)\delta_{nn'}$. Because the $u_{n\vec{k}\sigma}$ can be expanded in the $u_{n0\sigma}$, it follows that the average value of $|M_{n\vec{k}\sigma,n'\vec{k}'\sigma'}|^2$ equals the average value of $|M_{n0\sigma,n'0\sigma'}|^2$.

[18]$\Theta(p_0-\omega-E_F)=0$ for $p_0<E_F$ and $\Theta(p_0-\omega-E_F)=1$ for $p_0>E_F+\omega_{\max}$, where $\omega_{\max}$ is the largest value of $\omega_{\vec{q}}$.

[19]For Ni we would calculate $\Sigma^{\uparrow}$, the self-energy of majority spin holes, so that $dN^{\uparrow}/d\epsilon$ is replaced by $dN^{\downarrow}/d\epsilon$ evaluated over a region below E_F.

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[22]The [111] magnon with this wave vector has an energy of 0.083 eV but finite lifetime effects begin to be seen at 0.040 eV. This we believe (see Ref. 1) is much less than $E_F-E_d^{\uparrow}$ where $E_d^{\uparrow}$ is the energy at the top of the majority d bands. Thus we believe the finite magnon lifetime arises from the scattering of majority s and p electrons into the minority (mainly d) bands.