# Local-Band-Theory Analysis of Spin-Polarized, Angle-Resolved Photoemission Spectroscopy

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We present the local-band theory of the temperature dependence of up- and down-spin Green's functions in itinerant-electron ferromagnets. The results, calculated for a temperature-independent, local, exchange field strength, and under the assumption of the existence of short-range magnetic order, agree with recent measurements of angle-, energy-, and spin-resolved photoemission in nickel. States which contribute to the magnetization, not seen in the measurements, remain split in energy even at $T_C$. Split states at $T_C$ are also expected in iron.

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Angle-, energy-, and spin-resolved photoemission (ARPES) ideally is interpreted as the annihilation of a bulk electron of definite energy, momentum, and spin (and the creation of a high-energy electron which escapes and is measured). It measures the spectral density of the single-hole Green's function. The spin of this hole acts as a probe of the magnetic condition of the solid through which it propagates.

The beautiful experiments of Hopster *et al.*[1] using ARPES to study the magnetic properties of nickel have thus generated much interest, especially as it was claimed that no existing picture of high-temperature itenerant-electron magnetism is consistent with the results. It is our purpose here to show that the "local-band picture" *is* so consistent. It remains to be seen whether the "alloy analogy" picture is *also* consistent, as that theory has not been worked out for realistic bands.[2]

The local-band picture[3,4] stresses the existence of short-range magnetic order which remains on a scale of about 20 Å ($=2\pi/a$) above $T_C$. This allows spin-split energy bands to be quite well defined locally. The alloy analogy[5,6] idea is to neglect short-range order and assume atomically random mean spins, enabling an exploitation of the coherent potential approximation.

The question is to discuss the propagation of a hole starting with definite spin through two such different magnetic media. The results depend on the parameters of the hole state. There are two limiting cases. Namely, suppose the hole moves fast enough that the exchange field it sees changes direction in a time short compared to that which it takes its spin to align itself to the exchange field. The spin of such fast holes in effect sees the average magnetization, i.e., there is a *motional narrowing.* The spectral density will peak at different energies for spin up and spin down with the energy difference between peaks proportional to the magnetization. The spin of slow holes, on the other hand, follows the local magnetization and remains in approximate alignment or antialignment with it. There will be two peaks in the spectral density of each spin whose weights are coupled to the bulk magnetization, but which are split by the local magnetization.

Thus the critical parameter is $\hbar a v/\Delta$, where $\Delta$ is the spin splitting of the hole and $v$ its velocity. (The minimum value of $v$ is $\hbar a/m$, with $m$ the hole mass.) For nickel, the probing holes in the actual experiment have small mass and relatively small $\Delta$ leading to intermediate values of the hole speed. The magnetic electrons in nickel (which have only majority spins occupied) are significantly more massive and magnetically coupled, and are thus slower. In iron, with much larger $\Delta$, the slow approximation should be reasonable. In the alloy-analogy picture, the fast approximation is automatically made.

We have implemented these ideas with definite formulas. We first present the numerical results appropriate for the measurements in nickel. Figure 1 shows the theoretical temperature evolution for the measured,[1] intermediate-velocity, band states. These curves are for a local exchange field with the ground-state strength, and for a temperature-dependent scale of short-range magnetic order whose value in the paramagnetic state is what we estimat-

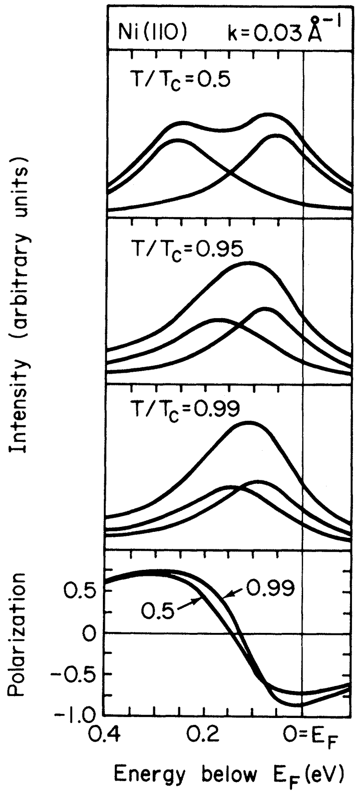

FIG. 1. Majority, minority, and total spectral function at three temperatures, for an $S_4$ symmetry band near the $X$ point in nickel. The three upper panels correspond to relative magnetizations $c = 0.90$, 0.45, and 0.25, respectively. The lower panel shows the polarization $(\uparrow - \downarrow)/(\uparrow + \downarrow)$, as a fraction of $c$, at the temperatures indicated.

ed earlier.[3] There is good agreement with the measured spectra,[1,7] after the substantial experimental and intrinsic broadening already present at low temperature is included. We find a different evolution for other bands, however, and for different states within a single band. Figure 2 shows a "magnetic" (partially singly occupied) band of nickel, just below $T_C$. The low-velocity states, which carry the magnetization, remain split. At higher velocity there is central-peak structure, as reported earlier[8,9] for *spin-unresolved* spectra on the (111) face. (We remark that this central peak is the sum of two spin-polarized peaks. The natural, but incorrect, assumption that the central peak was unpolarized led the authors of Ref. 1 to reject our earlier theory.[8] The present theory is consistent with the earlier one, but calculates the spin dependence, as well as giving a more sophisticated evaluation of the material parameters.) The difference between Figs. 1 and 2 is that the magnetic band has a higher effective mass and a larger ground-state splitting. The even larger splitting in iron should also give rise to well-separated peaks at $T_C$, as was recently reported.[10]

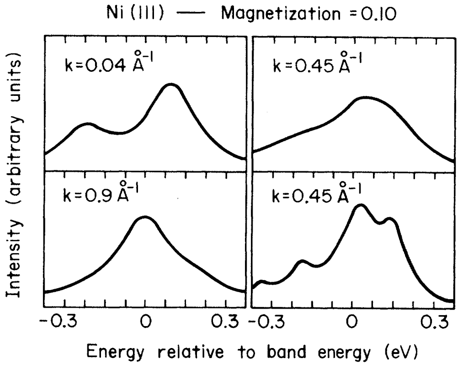

FIG. 2. Total spectral function just below $T_C$ for a "magnetic" band of nickel, at three values of initial momentum. The last panel has reduced intrinsic broadening, to emphasize the underlying structure.

We discuss next the low energy moments of the spectral functions. Consider the single-band Hamiltonian describing the *probe* electron,

$$H = \sum_{ij\sigma} T_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + (\Delta/2) \sum_{i\alpha\beta} c_{i\alpha}^{\dagger}\, \vec{\sigma}_{\alpha\beta} c_{i\beta} \cdot \hat{n}_i(t). \quad (1)$$

The exchange field is determined by the *magnetic* electrons and has strength $\Delta/2$, proportional to the *local* magnetization, and direction $\hat{n}_i(t)$. The paramagnetic band energies $\epsilon(k)$ are lattice Fourier transforms of $T_{ij}$.

We find the mean energies and low-order moments of the spin-up (-down) [+ (−)] spectral functions by manipulating the equations of motion[11]:

$$\bar{E}^{\pm}(k) = \epsilon(k) \mp c\Delta/2, \quad (2a)$$

$$\langle (E - \bar{E})^2 \rangle^{\pm}(k) = \Delta^2(1 - c^2)/4, \quad (2b)$$

$$\langle (E - \bar{E})^3 \rangle^{\pm}(k) = \pm \Delta^3 c(1 - c^2)/4 + \Delta^3 L(k)/4, \quad (2c)$$

where

$$\Delta L(k) = \langle (\hat{n}_i \cdot \hat{n}_j - 1) T_{ij} \rangle \pm \hat{z} \cdot \langle \hat{n}_i \times \partial \hat{n}_i / \partial t \rangle. \quad (2d)$$

In Eq. (2d), the angular brackets denote an average over the distribution of magnetization orientations appropriate for a given temperature. The quantity $c$ is $\hat{z} \cdot \langle \hat{n} \rangle$, so that $c\Delta$ is proportional to the bulk magnetization. $L(k)$, with similar terms for higher moments, measures the local directional disorder. Since this first appears in the third moment, it is apparent that the spectral function will not be as sensitive to the degree of short-range order as might intuitively have been expected.

If $L(k)=0$, then Eq. (2) describes a state of no disorder. The spectral functions then have peaks at $E=\epsilon(k)\mp\Delta/2$, with weights $(1\pm c)/2$ and $(1\mp c)/2$. This is the limit where local bands are precisely as in the ground state, and $\Delta$ has its ground-state value.

For large $L(k)$, the spin-resolved spectra may each show a single peak, as in Fig. 1. Then Eq. (2) implies a peak separation proportional to the net magnetization, while the peak widths increase until $T_C$, and remain constant above. This is the reported[1] behavior of the measured spectra. The broadening at $T_C$ is consistent with the ground-state value of $\Delta$, if intrinsic and magnetic-disorder broadening add in a mean-square sense. Then the local-band theory does describe the ARPES results correctly, even for converging peaks.

The magnetic bands (at $X$) do not collapse in this way, however. We can see why this is by estimating $L(k)$, which can be written

$$L(k)=\sum_q \chi(q)[\epsilon(k\mp q)-\epsilon(k)]/\Delta,$$

with $\chi$ the $\hat{n}\cdot\hat{n}$ correlation function. In a parabolic approximation for $\epsilon(k)$ this is $\langle q^2\rangle/m\Delta$. Using appropriate values for the bands involved, and our estimate[3] $\langle q^2\rangle\sim a^2$, we find $L$ of order unity at $T_C$ for Fig. 1, and 2.5 times smaller for Fig. 2. The short-range order is then just enough to prevent loss of the local magnetization, and the large energy cost that would entail.

To calculate in detail the spectral functions, we exploit the existence of short-range order by doing perturbation theory in gradients of $\hat{n}$. This is somewhat intricate, and details will be reported elsewhere. The Green's functions have the approximate form

$$G^{\pm}(k,E)=\frac{z-\Sigma_1-\Sigma_2\mp c(1-\Sigma_p)}{z^2-1-z(\Sigma_1+\Sigma_2)}. \tag{3}$$

In Eq. (3), $z=E-\epsilon(k)$, and $\Delta/2$ is the unit of energy. The self-energy functions $\Sigma_1$ and $\Sigma_2$ are those found earlier[9]:

$$\Sigma_{1,2}=\left(\frac{2}{2\pi}\right)\sum_q\int d\Omega\, F(q,\Omega)\left(V+\frac{V^2}{z^2\pm1-V}\right), \tag{4}$$

where the vertex $V=\epsilon(k\mp q)-\epsilon(k)\pm\Omega$, $\Sigma_p$ is $\Sigma_1/(z+1)+\Sigma_2/(z-1)$, and the function $F(q,\Omega)$ is defined below.

There are significant intrinsic broadening effects left out of our simple model, and substantial instrumental broadening in the measurements. We incorporate these by letting $z$ go to $z+i\gamma$, with $\gamma$ a phenomenological parameter chosen to mimic the observed widths at $T=T_C/2$.[12]

The Green's functions of Eqs. (3) and (4) satisfy important constraints which the exact functions must satisfy. They have the correct analyticity and positivity properties in the complex $z$ plane, and exactly satisfy the moment relations of Eq. (2), as well as the two next higher ones, for any given distribution of magnetization directions. In addition, they behave correctly[13] at low temperature, as compared with Fermi-liquid spin-wave theory.

The function $F(q,\Omega)$ is defined[9] so that $q^2F$ is the Fourier transform of the space- and time-dependent correlation function of certain gradients of $n$. There is no simple relation between $F$ and the corresponding $\chi(q,\Omega)$, but the $q$ moments of the equal time correlations are related as

$$\begin{gathered}4F^{[2]}=\chi^{[2]},\quad 4F^{[4]}=\chi^{[4]}-(\chi^{[2]})^2,\\ 4F^{[6]}=\chi^{[6]}-2(\chi^{[4]})(\chi^{[2]})+(\chi^{[2]})^3,\end{gathered} \tag{5}$$

where $F^{[n]}=(2\pi)^{-3}\int d^3q\, q^nF(q)$, and $\chi$ is normalized to $1-c^2$. These follow from the definition of $F$, and are necessary to satisfy the moment conditions of Eq. (2). We determine $\chi(q)$ from its normalization, given by the bulk magnetization, with $\Delta$ assumed constant, and from its $q^2$ moment, which is proportional to the magnetic energy.[1,14] Then Eq. (5) sets the parameters for $F(q)$. We take a spin-wave form for the frequency dependence of $F$, $F(q,\Omega)\propto\delta(\Omega-Dq^2)$.

Important contributions to the various moments of Eq. (5) come from larger values of $q$. To avoid overemphasizing poorly known high-$q$ fluctuations, we choose a flat spectrum for $\chi(q)$, cut off at finite wave vector $Q$. (At $T_C$, $Q\approx0.5$ Å$^{-1}$.) Similarly, $F(q)$ is taken as a sum of cut-off flat spectra, one somewhat narrower than $\chi$ and one very wide, but of small amplitude. Details of the computed line shape do depend on the form chosen for $F$, but the first three moments depend only on the magnetic energy and the magnetization.

For the magnetic energy we use a fit[15] near $T_C$, interpolated to a low-temperature spin-wave contribution. We use our estimate[3] that $\langle q^2\rangle^{1/2}=0.2$ Å$^{-1}$ well above $T_C$ in nickel, and scale by the estimated magnetic energy to find $\chi^{[2]}$ at a given temperature. Using an effective mass approximation for $\epsilon(k)$, and values of the mass and ground state $\Delta$ for the bands in question, we find the spectra of Figs. 1 and 2.

We have presented local-band-theory calculations for the temperature evolution of spectral functions for two bands of nickel. For one of these bands, the peaks converge at $T_C$, as seen experimentally. The magnetic band stays split, allowing substantial local magnetization in the paramagnetic state.

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