Unbiased Access to Exchange Splitting of Magnetic Bands Using the Maximum Entropy Method

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The experimental information about magnetic bands and their temperature dependence is distorted by Fermi distribution and apparatus function. The recovery of the spectral density constitutes an ill-posed inversion problem. We have invoked the maximum entropy regularization to deconvolute spin-resolved inverse photoemission data of Ni(110). The effective energy resolution is hereby improved by a factor of 5 and structures below E_F, which are generally lost in inverse photoemission, are recovered. The temperature-dependent data for Ni reveal clearly a pair of spin-split bands. The splitting vanishes on approaching the Curie temperature.

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The reconstruction, inversion, or deconvolution of experimental data is hampered very often by an ill-posed inversion problem. There exists an infinite manifold of possible solutions consistent with the experimental data within the error bars. A consistent probabilistic unbiased method to treat ill-posed problems is given by the maximum entropy (ME) concept [1,2] which is well established in the field of image reconstruction. In this Letter we show, guided by inverse photoemission (IPE) data for Ni, that ME can be equally successful in the analysis of spectroscopic data in solid state physics. In the case of IPE it allows one to improve the effective energy resolution by a factor of 5 and to recover structures of the spectral density below the Fermi energy which are generally lost. The same is true for the analysis of photoemission data with respect to structures above the Fermi level. This is important for the detailed understanding of the electronic structure close to the Fermi level, e.g., of high-temperature superconductors.

Here we present an application of ME to IPE data on nickel. The temperature and spin dependence of electronic states close to the Fermi level are important for the understanding of collective magnetism of itinerant electrons on a microscopic level. One topical question is to what extent the spin-dependent quasiparticle bands can be treated within a mean-field approximation, as it is assumed in state-of-the-art band-structure calculations, which leads to a Stoner-type exchange splitting between majority and minority bands. It is generally believed that ground-state and low-temperature properties of transition metals are fairly well described by the local density approximation, which yields one (Zeeman-shifted) band per spin direction. The high-temperature properties, however, are subject to controversy in the literature. Approximate many-body theories predict a multiband structure with temperature-dependent pole strength and quasiparticle energies. With increasing temperature, correlation effects are expected to lead to a mixing of spin-up and spin-down states resulting in "extraordinary" peaks. Above T_c the spin asymmetry disappears, as the rotational symmetry is restored. The multiband structure is, however, retained at and above T_c, due to short-range ferromagnetic spin correlations. These ideas are accounted for in, e.g., the fluctuating band theory [3] or approximate many-body calculations based on Hubbard-type model Hamiltonians [4] and cluster calculations [5]. At present there is, however, no theory for band magnetism which is generally accepted. Even the more fundamental question of whether model Hamiltonians, like the Hubbard model, describe ferromagnetism at all is still not settled [6]. It is therefore important to have accurate and conclusive experimental data to test various theories.

Experimentally, one has been trying to reveal the detailed spin and temperature dependence of the electronic states to discriminate between the mean-field (Stoner) picture and the multiband behavior. A number of PE and IPE studies on Fe and Ni dealt with either totally occupied or empty states which do not directly contribute to the magnetic moment. While for Fe clear evidence has been found for noncollapsing band behavior at specific points in **k** space [7], for Ni no clear-cut case has been observed so far [8], which has been attributed to the comparably small exchange splitting of ≤ 300 meV and the small amount of short-range magnetic order [3–5]. A partially filled and hence truly magnetic band has been studied on Ni(110) by spin-resolved IPE [9]. The investigated Z_2 band is a prototype of a narrow magnetic band which was expected to remain split even at T_c [3]. Model calculations have been performed for the two scenarios: (a) collapsing bands with one peak per spin direction and (b) two peaks per spin with temperature-dependent pole strength. Upon comparison with the experimental data strong evidence has been found in favor of the Stoner picture [9]. It could not, however, be ruled out that actually a mixture of (a) and (b) is present; neither has it been possible to determine the energetic positions of the bands close to E_F due to the limited experimental energy resolution of ≈ 450 meV.

To reveal detailed features of the spectral density, like quasiparticle energy and lifetime, particularly for states below the Fermi energy which are buried under the Fermi distribution, a more subtle analysis of the experimental data is required. To this end we invoked the ME method. The reasoning behind it is identical to that used

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in statistical physics to derive, e.g., the Fermi distribution function. Roughly speaking, among all possible solutions of the ill-posed problem, the one with maximum probability is taken. In many cases this is equivalent to the average of all solutions compatible with the experimental data within the error bars. It has been applied successfully to invert, reconstruct, or deconvolute incomplete and noisy data in a wide variety of physical applications, like radio and x-ray astronomy, Raman and NMR spectroscopies, tomography, analytic continuation of quantum Monte Carlo data, and image reconstruction [1,2]. ME provides a consistent description of probabilistic inference based on Bayesian statistics, the importance of which has been emphasized recently by Anderson [10].

The experimental IPE intensities for 100% spin-polarized electrons of spin σ are proportional to

$$I^{\sigma}(\omega,T,\mu)=j^{\sigma}\int A^{\sigma}(\omega')\{1-f(\omega',T,\mu)\}g(\omega'-\omega)\,d\omega'. \tag{1}$$

$A^{\sigma}(\omega)$ is the sought-for spectral density of quasiparticles with energy ω', spin σ, and wave vector $\mathbf{k}$ given by the experiment. j^{σ} represents the current density of incoming electrons of spin σ. To derive (1) standard approximations have been made, like ignoring matrix element and relaxation effects. In this approximation, the information about the electronic structure is entirely contained in the electronic spectral density. The dependency on temperature T and chemical potential μ enters via the Fermi distribution $f(\omega,T,\mu)$. $g(\omega'-\omega)$ stands for the apparatus function, a convolution of the energy distribution of the incoming electrons and the energy window for the detected photons. g can be estimated quite accurately from measurements of image-potential surface states on Ni(111) [11] which have small intrinsic linewidths. Figure 1 shows the first three members of the image-state series where the intrinsic linewidths have been taken from two-photon photoemission results [12]. We have added a linear background with a steplike increase at the vacuum level E_V [11], and convoluted the result with a Gaussian function of variable width σ. Comparing this simulation (solid line through data points) with the measured data (open circles) we find that the apparatus function can be approximated fairly well by a Gaussian with $\sigma=195$ meV (corresponding to FWHM $=460$ meV).

For numerical purposes we evaluate the spectral density $A_n^{\sigma}=A^{\sigma}(\omega_n)$ at discrete energies ω_n with $n=1, 2, \ldots, N_{\mathrm{var}}$ and interpolate it linearly in between. The integrals in Eq. (1) of the piecewise linear $A^{\sigma}(\omega)$ and the full Fermi function $f(\omega)$ are computed numerically. This procedure accounts for the rapid variation of $f(\omega)$ close to μ. Equation (1) transforms into a set of linear equations

$$I_l^{\sigma}=I^{\sigma}(\tilde{\omega}_l,T,\mu)=j^{\sigma}\sum_{i=1}^{N_{\mathrm{var}}}M_{li}A_i^{\sigma},\quad l=1,2,\ldots,N_{\mathrm{eq}}, \tag{2}$$

where $\tilde{\omega}_l$ represents the (coarser) mesh on which the experimental data are available. The spin polarization of the incoming electron beam has been estimated to $p=(N_{\uparrow}-N_{\downarrow})/(N_{\uparrow}+N_{\downarrow})\approx 33\%$ [9]. The incoming beam of predominantly spin-σ electrons contains, therefore, a percentage $n_{\sigma,\sigma}=(1+p)/2$ of spin-σ electrons, while the remaining percentage of electrons $n_{\sigma,-\sigma}=(1-p)/2$ has opposite spin. The measured intensities for σ-polarized beams contain a mixture of spectral densities for both spin directions and are proportional to

$$g_l^{\sigma}(\mathbf{A})=\sum_{\sigma',i}M_{li}n_{\sigma,\sigma'}A_i^{\sigma'}. \tag{3}$$

To recover the spectral density Eq. (3) has to be inverted. It appears that this inversion is utterly ill posed. In the first place, M_{li} is almost singular due to the Fermi function, which suppresses structures below the chemical potential ($|\omega-E_F|>k_BT$) exponentially. Consequently, the inverse matrix has very large eigenvalues and the experimental errors will be strongly amplified. The scatter of possible solutions, compatible with the experimental data, is therefore tremendous. A straightforward inversion of Eq. (3) (with $N_{\mathrm{var}}=N_{\mathrm{eq}}$) leads to results fluctuating between $+10^5$ and -10^5, while the real values for $A(\omega)$ are of order 1. Only if the experimental data would have a relative accuracy of better than 10^{-6} would the direct inversion of Eq. (3) be feasible. Moreover, this direct approach is restricted to ($N_{\mathrm{var}}\leq N_{\mathrm{eq}}$).

Using Bayesian statistics it is possible to determine the *posterior* probability $P(\mathbf{A}|\mathbf{g}^{\mathrm{exp}},\lambda)$ for a particular solution $\mathbf{A}$, given the experimental data $\mathbf{g}^{\mathrm{exp}}$ and additional experimental parameters λ, like scale of error bars, chemical potential, or width of experimental resolution:

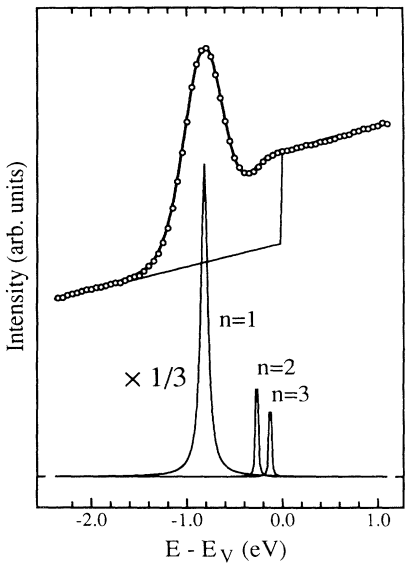

FIG. 1. Inverse photoemission data of the image-potential surface state on Ni(111) (open circles). A convolution of the first three members of the Rydberg series plus background with a Gaussian of $\sigma=195$ meV fits the experimental data well (solid line through the data points), which gives a good estimate of the apparatus function.

$$P(\mathbf{A}|\mathbf{g}^{\exp},\lambda)=P(\mathbf{g}^{\exp}|\mathbf{A},\lambda)\frac{P(\mathbf{A}|\lambda)}{P(\mathbf{g}^{\exp}|\lambda)}. \quad (4)$$

$P(\mathbf{g}|\mathbf{A},\lambda)$ is the *likelihood* function which contains the new information provided by the experiment. If the data are assumed independent and Gaussian distributed with error σ_i, the *likelihood* function is related to the χ-squared measure by

$$P(\mathbf{g}^{\exp}|\mathbf{A},\lambda)=e^{-\chi^2/2} \text{ with } \chi^2=\sum_{l=1}^{N_{eq}}\left[\frac{g_l^{\exp}-g_l(\mathbf{A})}{\sigma_l}\right]^2.$$

$g_l(\mathbf{A})$ is the predicted result for given $\mathbf{A}$ of Eq. (3). The *prior* probability $P(\mathbf{A}|\lambda)$ in Eq. (4) is determined by the information theory entropy S:

$$P(\mathbf{A}|\lambda)=e^{\alpha S} \text{ with } S=\sum_i A_i-m_i-A_i\ln(A_i/m_i).$$

m_i stands for the default model. We have chosen $m_i=\varepsilon$, where ε is a small quantity which merely serves to suppress noise in regions of insufficient information. The *posterior* probability is therefore given by

$$P(\mathbf{A}|\mathbf{g}^{\exp},\lambda)\propto e^{-\chi^2/2+\alpha S}. \quad (5)$$

The ME solution for $\mathbf{A}$ is obtained upon maximizing the *posterior* probability or rather $\alpha S-\frac{1}{2}\chi^2$. The regularization parameter α is determined self-consistently as elaborated by Skilling [13] upon maximizing the evidence $P(\alpha|\mathbf{g}^{\exp})$ for α, given the experimental data. Similarly other parameters, like the chemical potential, width of the Gaussian resolution, and degree of polarization, can be determined via the evidence analysis.

We have applied the ME deconvolution to temperature-dependent spin- and angle-resolved IPE data of Ni(110) for the $Z_4\rightarrow Z_2$ transition [9]. The experimental data are given for twenty energies per spin direction and $A_\sigma(\omega)$ is reconstructed for eighty energies. The inversion problem of Eq. (3) is therefore highly underdetermined. Experimental data have been available for temperatures $T/T_c=0.48$, 0.64, 0.72, 0.82, 0.95, and 1.02, covering the range of almost perfect ferromagnetic order into the paramagnetic regime. Before discussing the physical conclusions we will address characteristic parameters of the experiment: The error bars of the IPE data are given experimentally to $\leq 2\%$. ME analysis leads to a confirmation of these values, which is reasonable as the error bars in a counting experiment are known. Likewise for the chemical potential, only slight deviations $|\Delta\mu|\lesssim 0.04$ eV from the experimentally determined values were found. A further convincing result of ME concerns the apparatus function. We allowed more flexibility in the sense that the decay of the Gaussian can have different values for the left and right flank $\sigma_{l,r}$ of the peak. We find that the evidence is sharply peaked at $\sigma_l=183$ meV and $\sigma_r=195$ meV with a peak width of 1 meV. These values are in good agreement with our estimate given above and actually improve the agreement with the experimental data for the image states in Fig. 1. As the ME values for $\sigma_{l,r}$ have only very small uncertainty, this approach is very useful to determine the apparatus function for cases where it is not accessible by experimental means. Because of the incomplete spin polarization of the incoming beam one always observes two peaks in the experimental raw data. The polarization of the incoming beam had been determined experimentally such that the extraordinary peak vanishes for the $T/T_c=0.48$ data [9]. Using this experimentally determined value $p\approx 33\%$ for all temperatures we find almost negligible and temperature-independent "extraordinary" peaks. As ME is not a linear method it is expedient to give it the full experimental information in the form of Eq. (3) using p as the adjustable parameter subject to evidence analysis. As the polarization of the incoming beam is independent of the sample temperature, the same polarization p has been used for all temperatures and the combined evidence $\prod_i P(p,T_i)$ has to be maximized simultaneously. The maximum evidence is obtained for $p=32\%$, which is in good agreement with the experimentally determined value of $p=(33\pm 3)\%$. For this value of p extraordinary peaks disappear for all temperatures.

Typical results obtained by the ME deconvolution are given in Figs. 2(a) and 2(c) for $T/T_c=0.72$ and 0.82. For comparison we also depict the experimental data. In the experimental data both spin-up and spin-down features appear above E_F [9]. The reconstructed spectral densities, however, reveal the spin-up peak clearly below (above) E_F for $T/T_c=0.72$ (0.82) with a linewidth of about 80 meV, independent of temperature. The resolution of IPE+ME is better then 40 meV. The implicit reason is that the experimental resolution is due to a convolution with a smooth convolution function which can be characterized extremely accurately by a few parameters, independent of temperature. There is no significant indication for extraordinary peaks at all temperatures. To quantify this statement, we have determined the posterior

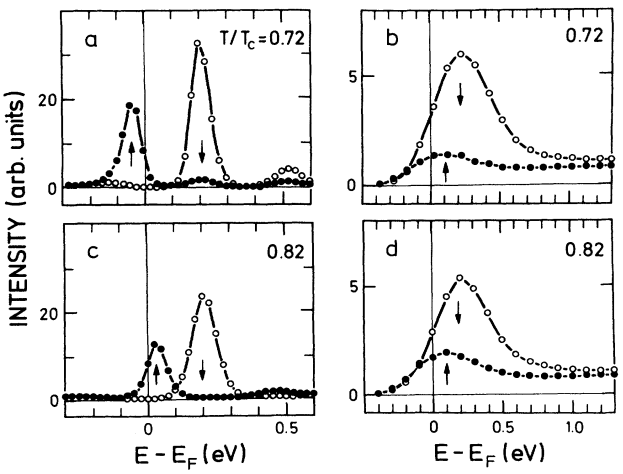

FIG. 2. Spin-dependent quasiparticle spectral density (a),(c) and experimental IPE data (b),(d) of the $Z_4\rightarrow Z_2$ transition in Ni for two temperatures, $T/T_c=0.72$ (a),(b) and 0.82 (c),(d).

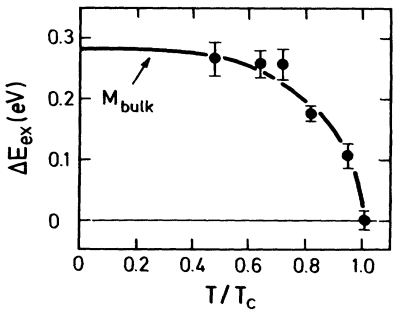

FIG. 3. Exchange splitting ΔE_{ex} of the Z_2 band in Ni as a function of temperature (full circles). Error bars are obtained self-consistently from ME. The full line is the experimental bulk magnetization curve of Ni rescaled to fit the ΔE_{ex} data [14].

probability of a two-peak structure, where we have mixed in a certain percentage q of the minority peak to the majority structure and vice versa. It appears that the posterior probability drops like $P(q)/P(0) \approx e^{-aq^2}$. Remarkably, for the $T/T_c = 0.72$ data of Fig. 2, the posterior probability reduces to $1/e$ already for $q = 0.009$ although the results are still within the experimental error bars. This emphasizes that extraordinary peaks can be ruled out. With increasing temperature a decreases slightly as the peaks approach each other. For $T/T_c = 0.95$, the posterior probability drops below $1/e$ for $q = 0.02$. The slight structures visible in Fig. 2 for $T/T_c = 0.72$ have to be attributed to noise and are completely absent for $T/T_c \geq 0.82$. The minor structure at ≈ 0.5 eV stems from irregularities in the experimental data some eV above E_F and has no physical relevance. It is worthwhile to compare the experimental data with those obtained by using the ME result for $A(\omega)$ in Eq. (3) [solid lines through the data points in Figs. 2(b) and 2(d)]. The agreement is perfect, but for ill-posed inversion problems this is not surprising. It is likewise a completely insensible test for theories to compare the theoretical and experimental values of g_l. The different heights of the peaks above and below the chemical potential should not be taken too seriously for the following reason. ME reduces structures in cases where the signal-to-noise ratio is poor, which is the case below μ due to the exponential decay of the Fermi function. The same argument leads also to a slight shift of structures in the direction where the kernel of the transformation increases. In the present case, we therefore expect that structures below μ are actually somewhat lower in energies. This effect is, however, accounted for in the error bars given by ME. The temperature dependence of ΔE_{ex} of the Z_2 band in nickel is given in Fig. 3. The data follow nicely the rescaled experimental bulk magnetization curve [14] which yields strong support for a Stoner-like band behavior. The extrapolated ground-state exchange splitting of the magnetic Z_2 band is 0.28 ± 0.05 eV. Similar values, ranging from 0.17 to 0.33 eV, have been reported for occupied d bands in Ni [15].

In conclusion, we have shown that the maximum entropy method allows one to infer spin-dependent quasiparticle spectral densities from IPE data. In the present case the resolution was improved by a factor of 5 and structures below E_F, which are generally lost in inverse photoemission, are recovered. We found that the quasiparticle spectral density consists of only one peak per spin direction for all temperatures. The exchange splitting ΔE_{ex} decreases with increasing temperature and vanishes at T_c. Hence, it appears that for the electronic bands in Ni the influence of transverse spin fluctuations is negligible. It has been shown that the ME concept is very useful to deconvolute experimental data. The procedure presented in this Letter can immediately be applied to other spectroscopies as well.

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