Evidence for Short-Range Magnetic Order in Ni above T_c

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A three-peak analysis of Ni(111) photoelectron spectra, taken with high angular and energy resolution at 7.7 eV for 300 K $\leqslant T \leqslant 1.25T_c$, is performed with the assumption of a temperature-independent exchange splitting and a many-body line shape. The intensity of the nonmagnetic peak between the two magnetic ones increases but its width decreases as T_c is approached from below, while the intensity of the magnetic peaks decreases but remains finite. This strongly supports the notion that short-range magnetic order persists well above T_c.

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Angle-resolved photoelectron spectroscopy (ARPES) started to be useful for the investigation of the magnetic properties of Ni when a doublet structure produced by the exchange splitting Δ_x of states at the top of the $3d$ band was identified in the (111) spectra at 300 K by three of the present authors.[1] Our results obtained for the photon energy $\omega = 10.2$ eV were later confirmed by more refined experiments.[2-5] It also became clear that a similar structure with a larger splitting observed in the spectrum[1] at $\omega = 16.8$ eV originates from two different states in the d band and not from the exchange splitting.[3] The intensity variation of the structure observed in the ARPES spectra with ω, which is very similar for Cu[6] and Ni,[7] shows that a single exchange-split $3d$ state is only obtained for $\omega < 13.5$ eV. The exchange splitting derived from ARPES agrees well with the value needed as a fit parameter in calculations reproducing the observed ω dependence of the photoelectron spin polarization,[8] while self-consistent calculations[9] give about twice that value. This discrepancy may be due to the inadequacy of the density-functional theory for strongly correlated systems as well as to final-state effects of the photoexcitation process.[10]

In this paper we present new ARPES spectra containing the exchange doublet for temperatures in the range 300 K $\leqslant T \leqslant 1.25T_c$, taken with a considerably higher resolution than that obtained in earlier experiments,[2,3] together with a new line-shape analysis. This enables us to attack directly the basic problem concerning the magnetism of Ni, namely, to find a model which adequately describes the curious mixture of itinerant and local features displayed by this metal. For example, the nonintegral magneton number and the existence of Δ_x are characteristic of a Stoner model, while the fact that $kT_c \ll \Delta_x$ contradicts it. On the other hand, the validity of the Curie-Weiss law for the susceptibility above T_c is characteristic of a Heisenberg model. To make matters worse, the existence of spin waves above T_c is alien to both models.[11,12]

The photoelectron spectrometer is basically the one described earlier,[13] except that we now use a high-intensity hollow-cathode H_2 discharge as the light source and a new electron mirror monochromator.[14] The angular and energy resolution of the spectrometer is 3° and 40 meV full width at half maximum for the spectra shown in Fig. 1. The geometrical configuration is shown as an inset. It corresponds to initial states with k vectors in the $(1\bar{1}0)$ mirror plane between the Λ and the Σ axis where the minority states close to the top of the d band are occupied.[1,4] The low photon energy of 7.7 eV and the fact that the radiation is polarized in a plane perpendicular to $(1\bar{1}0)$ assures that only these states contribute to the spectra shown in Fig. 1.[1,4]

We base our analysis on the local-band or short-range magnetic-order (SRMO) model which assumes an increase in transverse fluctuations of the spin-density vector with temperature but no change of its magnitude.[15,16] Above T_c these fluctuations destroy the long-range magnetic order, while the SRMO on a microscopic scale persists. According to Korenman and Prange,[17] the photoelectron spectra at elevated temperatures should then consist of two magnetic peaks separated es-

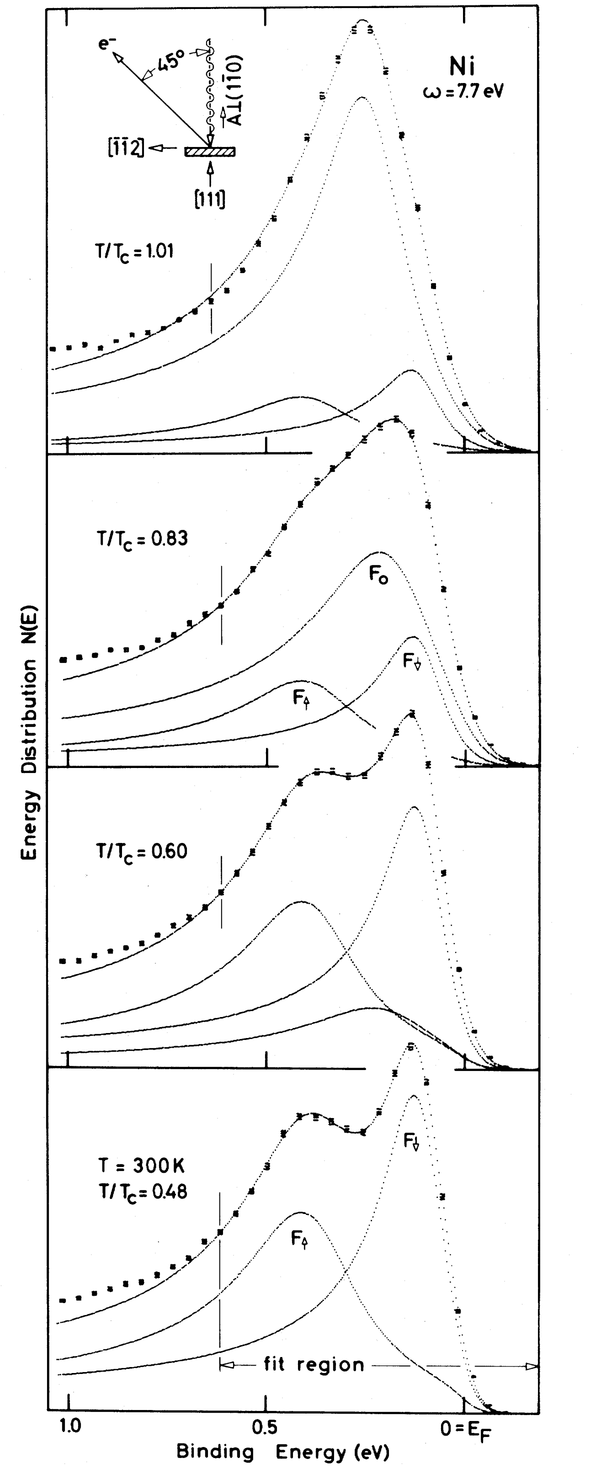

FIG. 1. Angle-resolved Ni(111) photoelectron spectra for $\omega = 7.7$ eV at four different temperatures. The detection geometry is shown in the inset. The measured points are plotted as rectangles; the height of these rectangles is twice the statistical error. The finely dotted curves are obtained by a least-squares fit (see text).

sentially by the same Δ_x as for low temperatures; in addition a "nonmagnetic" peak should develop in between the two magnetic lines, with its intensity increasing and its width decreasing as T_c is approached from lower temperatures. The decrease in width corresponds to the increase in the transverse fluctuations of the spin-density vector, i.e., to scattering of the photoelectrons at an increasingly nonmagnetic potential; it is similar to the "motional narrowing" observed in nuclear magnetic resonance.

Deviating from the usual practice in ARPES,[2,3] we use the many-body Doniach-Sunjić[18] (DS) line shape in our data analysis. This seems reasonable since the excitation from the relatively flat $3d$ band should produce similar low-energy electron-hole pairs as the excitation from core states. The assumption of a constant electron-hole excitation spectrum inherent in the DS formulation is of course not valid here. In order to approximate it better, we restrict the energy region of analysis as shown in Fig. 1. This also serves to minimize the influence of extrinsic losses contributing to the high-binding-energy side of the spectrum. In contrast to the x-ray photoemission case there is no need to fold a spectrometer function into the DS lines since the energy window of our spectrometer is much smaller than the linewidths encountered. A DS line is given by the expression

$$F_i(E) \propto \frac{\cos\{\pi\alpha/2 - (1-\alpha)\arctan[(E-E_i)/\gamma_i]\}}{[(E-E_i)^2 + \gamma_i{}^2]^{(1-\alpha)/2}}.$$

This expression goes over into $1/(E-E_i)^{1-\alpha}$ for vanishing lifetime width $2\gamma_i$ and into a Lorentzian for $\alpha \to 0$. At room temperature we perform a least-squares–fit decomposition of the ARPES spectrum into two DS lines with a common asymmetry factor α, i.e., we write $N(E) = C(F_\uparrow + F_\downarrow)f$, where f is the Fermi-Dirac function. This involves variation of the seven parameters C, $E_\uparrow$, $E_\downarrow$, $\gamma_\uparrow$, $\gamma_\downarrow$, α, and the Fermi energy E_F. The fit yields $\alpha = 0.225$ and $E_\uparrow - E_\downarrow = 0.271$ eV which converts to $\Delta_x = 0.30$ eV, by incorporating the finite slope of the initial band, in good agreement with the Δ_x values given earlier.[1-5] In the spirit of the SRMO model we assume the spectra for $T > 300$ K to consist of two magnetic components, taken to be the 300-K DS lines, and a third nonmagnetic component F_0, also assumed to be of the DS form with the same α, i.e., $N(E) = C[(F_\uparrow + F_\downarrow)(1-\eta_0)/2 + \eta_0 F_0]f$. In this high-temperature least-squares fit, the five parameters C, E_0, γ_0, η_0, and ϵ are varied, where ϵ is a temperatureinduced shift of the experimental energy scale (0–30 meV). Figure 1, which shows a selection from the fourteen spectra taken at various temperatures, gives the calculated $N(E)$ as the dotted lines together with the decomposition into the DS lines.

The most important results of the fitting procedure are condensed in Fig. 2. It does indeed show the expected increase in the fractional intensity η_0 of the nonmagnetic component F_0, and even more significantly, also the motional narrowing with increasing temperature as predicted by Korenman and Prange.[17]

Furthermore, it turns out that the *square* of the reduced saturation magnetization coincides with the fractional intensity of the magnetic components, i.e., $(M_s/M_{s0})^2 = 1 - \eta_0$, for $T \leq 0.9T_c$ (see Fig. 2). It is thus tempting to think of the photoemission experiment as a scattering experiment, with the scattering *amplitude* of the photoelectrons contributing to the magnetic components in proportion to the saturation magnetization, although we realize that the actual problem is considerably more complicated. The observed relation is also reasonable since the macroscopic magnetization and the magnetization of a microscopic region sampled by the photoelectrons are expected to be identical for the temperature region below T_c. The size of the microscopic region is given by the escape depth of the photoelectrons which is approximately 20 Å for photoelectrons of about 7.5 eV above E_F, typical for the experiment discussed here.[19,20]

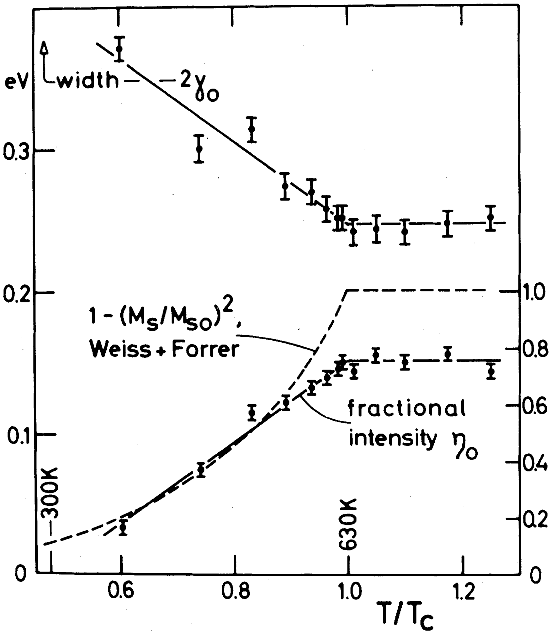

FIG. 2. The fractional intensity η_0 and the lifetime width $2\gamma_0$ of the nonmagnetic component F_0 as a function of the reduced temperature. The straight lines are obtained from the data points by another least-squares fit. The dashed line is 1 minus the square of the reduced saturation magnetization.

In contrast, for the temperature region above T_c, where the macroscopic magnetization is zero, a nonzero local magnetization of the 20-Å region sampled by the photoelectrons persists, as the value $1 - \eta_0 \cong 0.24$ for the fractional intensity of the magnetic components indicates (see Fig. 2), again in agreement with the SRMO model. In fact, by defining $\cos^2\bar{\theta} = 1 - \eta_0$, we obtain $\bar{\theta} \cong 60°$ as the average tilt angle of the spin-density vector with respect to the average magnetization direction of a 20-Å region for T above T_c. The measurements also indicate that the strength of the transverse fluctuations of the spin-density vector no longer depends on T for $T_c \leq T \leq 1.25T_c$ since neither the fractional intensities nor the width of the nonmagnetic component changes in this region.

It is also possible to fit the high-temperature ARPES spectra with two DS lines using Δ_x as an adjustable parameter. By equating the reduced exchange splitting with the reduced magnetization at 300 K, we obtain $\Delta_x/\Delta_{x0} \cong M_s/M_{s0}$ below T_c and $\Delta_x/\Delta_{x0} \cong 0.4$ above T_c. This is to be compared with $\Delta_x/\Delta_{x0} \cong 0.6$ as obtained by Eastman *et al.* who assumed no change of the lifetime width with temperature.[2] We reanalyzed their data, incorporating the strong energy dependence of the width by linearly interpolating between the 300-K values, and found $\Delta_x/\Delta_{x0} \cong 0.4$, in agreement with the two-peak analysis of our spectra which treats the width as fit parameters. The nonzero value of Δ_x above T_c has been taken as evidence for a modified Stoner model, characterized by the relation $\Delta_x/\Delta_{x0} = M^{\mathrm{loc}}/M_0^{\mathrm{loc}}$, where the local or microscopic magnetization sampled by the photoelectrons has replaced the macroscopic magnetization of the original Stoner model.[2] However, Korenman and Prange have already shown that this assumption is not justified.[17] Even on such a microscopic scale the Stoner-Hartree-Fock theory is not valid and the full scattering of the electrons from the spin fluctuations has to be taken into account. By generalizing their analysis it can be shown that a three-peak structure is obtained irrespective of the amount of SRMO, with only the weight of the magnetic peaks depending on the amount of SRMO.[21] This result justifies our evaluation above that the weight $1 - \eta_0$ of the magnetic peaks instead of the peak separation is a measure of the microscopic or local magnetization.

In summary, with the assumption of a temperature-independent Δ_x and the many-body DS line shape, the three-peak analysis of our spectra does indeed yield the temperature dependence of the intensities and the motional narrowing predicted by the SRMO model. The intensity of the two magnetic components and thus the degree of SRMO remains finite well above T_c.

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[1] E. Dietz, U. Gerhardt, and C. J. Maetz, Phys. Rev. Lett. 40, 892 (1978).

[2] D. E. Eastman, F. J. Himpsel, and J. A. Knapp, Phys. Rev. Lett. 40, 1514 (1978).

[3] F. J. Himpsel, J. A. Knapp, and D. E. Eastman, Phys. Rev. B 19, 2919 (1979).

[4] U. Gerhardt, C. J. Maetz, A. Schütz, and E. Dietz, J. Magn. Magn. Mater. 15–18, 1141 (1980).

[5] P. Heimann and H. Neddermeyer, J. Magn. Magn. Mater. 15–18, 1143 (1980).

[6] E. Dietz and D. E. Eastman, Phys. Rev. Lett. 41, 1674 (1978).

[7] E. Dietz, unpublished.

[8] E. Kisker, W. Gudat, M. Campagna, E. Kuhlmann, and I. D. Moore, Phys. Rev. Lett. 43, 966 (1979).

[9] C. S. Wang and J. Callaway, Phys. Rev. B 15, 298 (1977).

[10] A. Liebsch, Phys. Rev. Lett. 43, 1431 (1979).

[11] H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. 30, 556 (1973).

[12] J. W. Lynn and H. A. Mook, Phys. Rev. B 23, 198 (1981).

[13] E. Dietz and U. Gerhardt, J. Phys. F 8, 2213 (1978).

[14] C. J. Maetz, U. Gerhardt, E. Dietz, A. Ziegler, and R. J. Jelitto, to be published.

[15] V. Korenman, J. Murray, and R. Prange, Phys. Rev. B 16, 4032, 4048, 4058 (1977), and 19, 4691, 4698 (1979).

[16] H. Capellmann, Z. Phys. B 34, 29 (1979), and 35, 269 (1979).

[17] V. Korenman and R. E. Prange, Phys. Rev. Lett. 44, 1291 (1980).

[18] S. Doniach and M. Šunjić, J. Phys. C 3, 285 (1970).

[19] D. E. Eastman, Solid State Commun. 8, 41 (1970).

[20] J. W. T. Ridgeway and D. Haneman, Surf. Sci. 24, 451 (1971).

[21] E. Haines, V. Heine, and A. Ziegler, to be published.