Temperature Dependence of the Exchange Splitting in Ni Studied by Spin-Polarized Photoemission

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(Received 2 June 1983)

Angle- and spin-resolved photoemission spectra from Ni(110) have been measured in the temperature range $0.5 \leq T/T_C \leq 0.94$. The data cannot be reconciled either with the predictions of local band theory assuming a temperature-independent exchange splitting or with a pure Stoner model. It is concluded that the exchange splitting decreases with increasing temperature and that spin fluctuations strongly influence the photoemission line shapes.

PACS numbers: 75.50.Cc, 75.40.-s, 79.60.Cn

The ground state of the ferromagnetic 3*d* metals (Fe, Co, Ni)—with their nonintegral numbers of Bohr magnetons per atom—can be described by the Stoner model of band ferromagnetism. The spin-up (majority) and spin-down (minority) states are shifted with respect to each other by the exchange splitting Δ_{ex}. The appropriate description of the state at *finite* temperature is a matter of great current interest. In the Stoner model the exchange splitting decreases to zero at the Curie temperature T_C yielding the paramagnetic band structure. An opposite point of view is taken by local band theories.[1-6] In these theories, which were motivated by the observation of spin waves above T_C by inelastic neutron scattering,[7] short-range magnetic order is assumed, allowing the definition of a local ferromagnetic band structure. The reduction of the macroscopic magnetization with increasing temperature is then due to *transverse* fluctuations of the microscopic regions which retain essentially their $T=0$ magnetization. The exact nature and the spatial extent of the magnetic short-range order is a matter of current debate.[8,9]

Angle-resolved photoemission spectroscopy (ARPES) has been used to study spin-split electronic states in Ni. Upon approaching T_C an overall narrowing of the energy distribution curve (EDC) was found and the spin-split peaks could no longer be resolved. Very different conclusions were reached depending on the curve fitting procedures used. Eastman, Himpsel, and Knapp[10] interpreted their data as showing a reduction of the exchange splitting with increasing temperature of about 40%. On the other hand, it was shown by Maetz *et al.*[11] that the data are also consistent with a prediction of local band theory based on a temperature-independent exchange splitting.[12]

Recently it was shown that spin-polarized angle-resolved photoelectron spectroscopy (SPARPES) is now possible with a resolution comparable to conventional ARPES.[13] In this Letter we present the first temperature-dependent angle- and spin-resolved photoemission spectra. We have measured the photoemission spectra for normal electron emission from a Ni(110) surface using linearly polarized Ne I (16.85 eV) radiation from a resonance lamp. The light polarization for this study was chosen ($\vec{A} \| [110]$) so that only bands of S_4 symmetry at the X point contribute to the emission. At this point of the Brillouin zone both the spin-up and spin-down bands are located just below the Fermi energy. A more detailed discussion and a description of the experimental procedure is given elsewhere.[13] The sample, a picture-frame single crystal, was heated by radiation from a bifilar tungsten filament. No disturbing effect of the magnetic field of the heating current was observed. The temperature was measured by a NiCr/Ni thermocouple spot welded to the sample. The temperature reading was accurate within 3°. As a result of the decrease of the spin polarization at elevated temperatures, increasing measuring times were needed in order to obtain comparable relative statistical accuracy for the spin polarization. To ensure surface cleanliness the measurements were periodically interrupted for sputtering and flashing cycles.

In Fig. 1 we show the total EDC's and the spin-

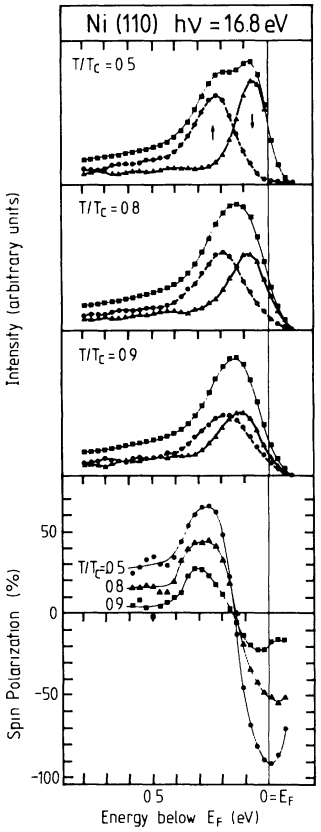

FIG. 1. Upper part: Total energy distribution curves (EDC's), $I_0 \propto I_\uparrow + I_\downarrow$ (squares), and spin-resolved EDC's $I_\uparrow = I_0(1+P)/2$ (circles), $I_\downarrow = I(1-P)/2$ (triangles), for three different temperatures; lower part: corresponding spin polarization curves, $P = (I_\uparrow - I_\downarrow)/(I_\uparrow + I_\downarrow)$.

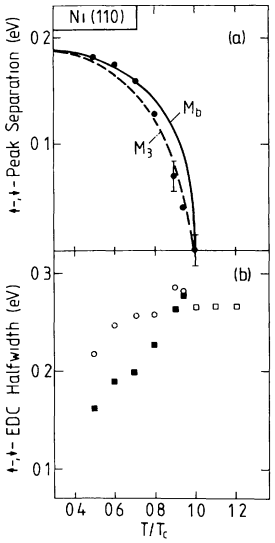

FIG. 2. (a) Energy separation of the spin-up and spin-down peaks as a function of temperature; the solid and broken lines represent the temperature dependence of the bulk magnetization and the magnetization of the third atomic layer, respectively; (b) half-width (full width at half maximum) of the spin-resolved energy distribution curves: circles, majority peak; full squares, minority peak. Above T_C (open squares) the spin-resolved EDC's are identical.

resolved EDC's for three selected temperatures as well as the corresponding energy-resolved spin polarization. First, we notice the *narrowing of the total EDC's* with increasing temperature and the disappearence of the double-peak structure in accordance with the ARPES data.[10,11] Second, we see that the *spin-resolved EDC's* approach each other with increasing temperature—Fig. 2(a) shows the peak separation—and become *broader*—Fig. 2(b) shows the half-width.

In this experiment the measured spin polarization is a macroscopically averaged quantity (over the diameter of the light spot, i.e., ~ 2 mm). This implies that above T_C the measured spin polarization is zero. In the energy distribution curves, on the other hand, there might still be structures due to short-range-order effects, like in the interpretation by Maetz *et al.*[11] There are two clear-cut theoretical predictions for the temperature dependence of the photoemission line shapes. First, the Stoner model predicts a decrease of the peak separation of the spin-resolved EDC's proportional to the macroscopic magnetization. The data of Fig. 2(a) are consistent with this. Actually the peak separation decreases slightly faster than the bulk magnetization. It follows more closely the temperature dependence of the magnetization of the third or fourth atomic layer in a mean-field calculation. This is expected, since the mean probing depth in this experiment is only about 3.5 atomic layers. The large broadening shown in Fig. 2(b), on the other hand, contradicts a simple Stoner behavior. We think that the broadening is mainly of magnetic origin and that phonon broadening is of minor importance. This is corroborated by the fact that above T_C the total EDC no longer changes [Fig. 2(b)].

The second prediction concerning the photoemission line shapes comes from local band theory.[12] Since we measure electron states at the boundary of the Brillouin zone (around the X point) the group velocity is nearly zero. For this case local band theory predicts a temperature-independent EDC and the spin polarization should scale down with the temperature dependence of the magnetization by a factor $M(T)/M(T=0)$ because

of the transverse fluctuations of the microscopic regions still having the full $T=0$ exchange splitting. That this prediction does not hold true can already be seen from the narrowing of the total EDC. It could be argued that because of the small escape depth and the resulting k smearing considerable intensity is originating from k vectors around the X point where the group velocity is not negligible. For this situation the local band theory also predicts a narrowing of the EDC due to the growth of the so-called nonmagnetic peak in the middle between the two magnetic peaks which stay at their low-temperature position and decrease in intensity. To investigate this possibility we have performed a three-peak fit of the EDC, as was performed in Ref. 11. The fit consists of an up-spin peak $I_\uparrow$, a down- spin peak $I_\downarrow$, and an unpolarized peak I_0 between $I_\uparrow$ and $I_\downarrow$. For the $I_\uparrow$ and $I_\downarrow$ we took the measured spin-resolved EDC's at room temperature. From this fit we can calculate the spin polarization $P=(I_\uparrow-I_\downarrow)/(I_\uparrow+I_\downarrow+I_0)$. This is of course an *overestimate* since the up and down directions refer to the local magnetization axis whereas the polarization is measured with respect to the macroscopic magnetization axis so that transverse fluctuations can only *reduce* the measured polarization. Despite this fact the measured polarization is already *higher* (by about 30–40%) than the maximum predicted by this three-peak fit. This shows that our data cannot be reconciled with the three-peak structure analysis based on a temperature-independent exchange splitting.[14]

Although the measured separation of the spin-resolved EDC's decreases to zero at T_C [Fig. 2(a)] this does not necessarily imply that locally Δ_{ex} is zero, because transverse fluctuations can reduce the measured polarization. If we perform a two-peak fit of the EDC at T_C, i.e., by taking the two spin-resolved EDC's measured at room temperature and letting them approach each other, we arrive at a reduction of the exchange splitting of about 50%, in agreement with the values obtained by Eastman *et al.* and also Maetz *et al.* for their two-peak analysis. The observed broadening of the measured *spin-resolved EDC's* can then be due to transverse fluctuations.

We note also that our results are in qualitative agreement with the probability distribution function for the local magnetic moments of Ni above T_C given by Hubbard[15] if we interpret this as a distribution of exchange splittings. He found an average 25% decrease of the $T=0$ local moment and a strongly asymmetric distribution extending towards small moments. Additionally, transverse fluctuations of these moments could lead to a broadening of the spin-resolved EDC's. In a recent calculation a reduction of the local moment of 50% has been found.[16]

Usami and Moriya[17] gave a theoretical prediction for the spin-unresolved photoemission line shapes within their spin fluctuation model (for a quantitative comparison with the present data more detailed calculations are needed). In their curves it seems, however, that the narrowing of the EDC continues beyond T_C in contrast to experiment. The experimental data seem therefore to imply that at T_C Ni has reached a state of magnetic disorder such that it is not affected significantly by further increase of the temperature.

In conclusion, we have shown that the new data of temperature-dependent spin-polarized angle-resolved photoelectron spectroscopy (SPARPES) for Ni are inconsistent with a temperature-independent exchange splitting as assumed in local band theory based on strong short-range magnetic order. Indications for the importance of longitudinal and transverse spin fluctuations have been found in the observed broadening of the spin-resolved energy distribution curves. Quantitative theories are now needed to explain the experimentally observed line shapes. The present SPARPES measurements are only a beginning and future experiments on materials exhibiting more localized or more itinerant behavior than Ni will contribute to our understanding of the interplay between changes of the electronic structure and magnetic phase transitions.

We thank many colleagues for helpful and stimulating discussions and comments during the workshop on 3*d* metallic magnetism at Institut Laue-Langevin, Grenoble, 25–26 March 1983. Special thanks are due to Professor H. Capellmann, Professor D. M. Edwards, Professor U. Gerhardt, Professor V. Heine, Professor R. E. Prange, and Professor E. P. Wohlfahrt. The experimental part of this work was performed at the Institut für Festkörperforschung of the Kernforschungsanlage Jülich and supported by the Deutsche Forschungsgemeinschaft through Sonderforschungsbereich 125.

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