# Temperature Dependence of the Exchange Splitting of Fe by Spin-Resolved Photoemission Spectroscopy with Synchrotron Radiation

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Temperature-induced changes in the electronic structure of Fe(100) have been investigated by spin- and angle-resolved photoemission for temperatures between room temperature and the Curie temperature $T_C$. States nearly stationary in energy ($\Gamma_{25}'^{\downarrow}, \Gamma_{12}^{\uparrow}$) have been observed for photon energy $h\nu = 60$ eV. However, from a strong increase in minority-spin intensity for $h\nu = 31$ and 21 eV, a downwards shift of the $\Delta_5^{\downarrow}$ band is inferred to occur upon heating towards $T_C$ for large $k$ vectors.

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The electronic structure at finite temperatures of the $3d$-transition metals Fe, Co, and Ni is currently a matter of strong theoretical interest. Spin-polarized band theory based on the self-consistent local-density-functional description gives an adequate account of the ferromagnetic ground state (e.g., cohesive energy, nonintegral moments).[1] However, controversial attempts have been made recently to describe transition-metal magnetism at finite temperatures.[2-6] The basic common idea is to try to incorporate into the theory the existence of local magnetic moments even above $T_C$. The ferromagnetic-to-paramagnetic phase transition is then governed by thermal disordering of the moments, requiring much less energy than single-particle spin flips which would involve energy changes as large as the exchange splitting. The controversy is over the spatial extent of correlation among the magnetic moments, which is connected intimately to the present debate on the existence of spin waves above $T_C$.[7]

The ferromagnetic-to-paramagnetic phase transition of Fe has been studied by spin-unresolved, angle-resolved photoemission.[8] However, only by measuring the electron spin explicitly can exchange-split bands be identified unambiguously and the band dispersions be detected, as will be shown below. Furthermore, the spin dynamics at elevated temperatures, as spin rotations around the spontaneous magnetization direction or flips of local magnetic moments which currently are considered to be the driving force for the phase transition, can be observed only by this method. We have therefore, for the first time, performed a spin-, angle-, and energy-resolved photoemission experiment on temperature-induced changes in the electronic structure of Fe. Because of a predicted wave-vector ($\vec{k}$) dependence of the temperature dependence of the exchange splitting,[5,9] we employed monochromatized tunable synchrotron radiation from the German storage ring BESSY, allowing selection of initial states with different $\vec{k}$.

The experiment is similar to a recent one on Ni(110)[10] using a resonance lamp. Total energy resolution, including the linewidth of light, was 0.4 eV at $h\nu = 60$ eV and about 0.3 eV at $h\nu = 31$ eV. The angular resolution was about $\pm 3°$ at $h\nu = 60$ eV decreasing to about $\pm 4°$ at $h\nu = 31$ eV, resulting in $k$ resolution of about $\frac{1}{5}$ of the Brillouin zone. The sample was cleaned *in situ* by standard surface-analysis techniques and its surface conditions were monitored by low-energy electron diffraction and photoelectron spectroscopy.[11] The sample was mounted with the easy magnetization direction (001) parallel to the spin-polarization-sensitive axis of the Mott (spin) analyzer and was magnetized in this direction. From the left and right counting rates $I_1$ and $I_2$ of the Mott detector the spin-resolved energy distribution curves (SREDC's) $I^{\uparrow}(E)$ and $I^{\downarrow}(E)$ are obtained as

$$I^{\uparrow} = 0.5[I + (I_1 - I_2)/S],$$
$$I^{\downarrow} = I - I^{\uparrow} = 0.5[I - (I_1 - I_2)/S]. \quad (1)$$

$S$ (=0.18) is the value of the foil-thickness-corrected Sherman function. $I$ is the spin-summed counting rate. The SREDC's are normalized to the light intensity at any selected photon energy to detect the transfer of electrons of one spin state into the other one occurring possibly at elevated temperatures. Further experimental details will be published elsewhere.[11]

As a result of the loss of spontaneous magnetization above $T_C$, the SREDC's become equal at and above $T_C$. The information on the changes in the microscopic electronic structure is contained in the way they approach each other.

Dipole selection rules indicate that only $\Delta_5$ symmetry bands along the $\Gamma$-$H$ direction (see Fig. 1) are allowed as initial states for the Fe(100) surface

 

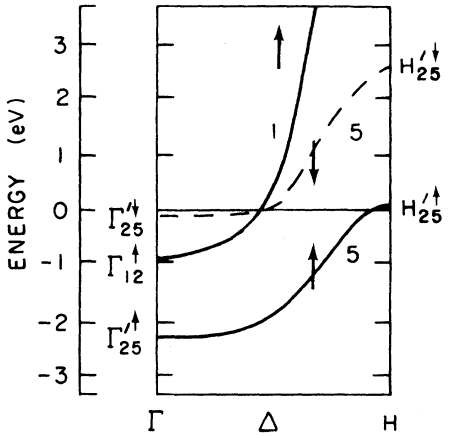


FIG. 1. The Fe Γ-$H$ direction of the band structure (Ref. 12) which is sampled by the present experiment.

with $s$-polarized light and normal emission. The band dispersions, like the minority-spin peak position, shift towards the Fermi energy ($E_F$) and, once it has reached $E_F$, the strong drop in intensity has been followed by tuning the photon energy from 60 to 20 eV; see Fig. 2.[13] Below 35 eV, the minority (↓) transition apparently takes place via indirect transitions from states where the $\Delta_5^{\downarrow}$ band crosses $E_F$ as concluded from the minor influence on photon energy between 31 and 20 eV. The majority (↑) SREDC's display the dispersion of the $\Delta_5^{\uparrow}$ band, its high-energy peak moving towards $E_F$ from $h\nu = 31$ to 21 eV corresponding probably to direct transitions from initial states in the right half of the Brillouin zone.

SREDC's taken at $h\nu = 60$ eV are shown in Fig. 3 for two different temperatures. In the $T/T_C = 0.3$ data, we identify the peaks in accordance with Feder *et al.*[14]: the ↓-SREDC displays only one single sharp peak ($\Gamma'^{\downarrow}_{25}$) at 0.4 + 0.2 eV below $E_F$. In the ↑ SREDC two peaks are resolved, one located at a binding energy $E_B = 2.6 + 0.2$ eV ($\Gamma'^{\uparrow}_{25}$) and the other one at 1.2 + 0.2 eV ($\Gamma^{\uparrow}_{12}$). The peaks at $E_B = 2.6$ and 0.4 eV are due to emission from the exchange-split $\Delta_5^{\uparrow}, \Delta_5^{\downarrow}$-symmetry bands. Emission from $\Gamma^{\uparrow}_{12}$ actually is forbidden for strictly normal emission.

Upon heating to $T/T_C = 0.85$ the following changes are observed (see Fig. 3): In the minority SREDC, the peak $\Gamma'^{\downarrow}_{25}$ diminishes strongly in intensity while its energy width increases by about a factor of 3, and its peak position shifts by 0.2 eV to larger binding enery. A reason for the intensity loss is an increase in angular width of the ↓-electron emission cone, measured by sweeping the beam across an aperture.[11] This leads to a smaller detected intensity since the angular acceptance of the apparatus is fixed. At $E_B = 2.6$ eV a new, broad peak emerges. In the ↑ SREDC, the $\Gamma'^{\uparrow}_{25}$ peak at $E_B = 2.6$ eV loses intensity, but much less than its exchange-split counterpart. Its position remains nearly unchanged. A small ↑-spin intensity gain is observed around the binding energy of $\Gamma'^{\downarrow}_{25}$.

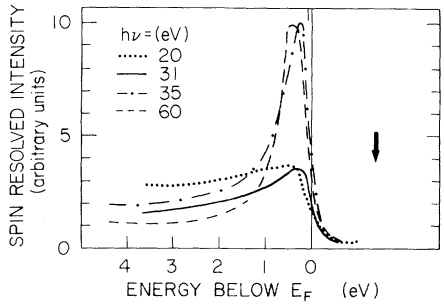


FIG. 2. Minority-spin energy-distribution curves from Fe(100) for normal emission and $s$-polarized light at different photon energies. The data are, at any photon energy, normalized to the maximum of ↑- or ↓-spin EDC, whichever is greater.

A marked feature of Fig. 3 is that the binding energy where the ↑ and ↓ SREDC's cross each other is the same for $T/T_C = 0.3$ and $T/T_C = 0.85$ (and also at intermediate temperatures). At this particular energy no net transfer of one spin state into the

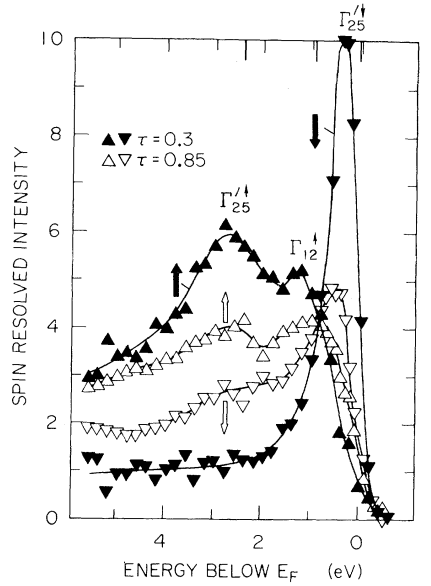


FIG. 3. Spin- and angle-resolved energy-distribution curves of Fe(100) taken at 60-eV photon energy for two different temperatures, $\tau = T/T_C = 0.3$ and 0.85, for normal emission and $s$-polarized light (unsmoothed data). The arrows labeling the curves refer to the direction of the spontaneous magnetization.

other one occurs upon heating.

Spin-summed EDC's show more clearly than Fig. 3 that around the $\Gamma_{25}'^{\uparrow}$ peak position the *total* intensity does not change between $T/T_C = 0.3$ and $T/T_C = 0.85$, indicating that losses in the $\uparrow$ SREDC are compensated for (to within $\sim 5\%$) by the new growing peak in the $\downarrow$ SREDC. This is not the case at the position of the $\Gamma_{25}'^{\downarrow}$ peak, where the total intensity drops upon heating because of the stronger losses in the $\downarrow$ SREDC as compared to the gains in the $\uparrow$ SREDC. The decrease in intensity at $\Gamma_{25}'^{\downarrow}$ is also larger than expected from the Fermi-Dirac function. We note that the intensity recovers upon cooling indicating that the observed changes are not related to contamination.

In the EDC taken at $T = T_C$ it is observed that the $\Gamma_{25}'^{\uparrow}$ peak remains stationary in energy (a possible shift towards $E_F$ is[11] less than 0.5 eV). The peak due to $\Gamma_{25}'^{\downarrow}$ apparently is smeared out under the stationary $\Gamma_{12}^{\uparrow}$ peak.

The balancing of gains and losses in intensity observed at the position of $\Gamma_{25}'^{\uparrow}$ could be interpreted in terms of fluctuations of a constant magnetic moment around the spontaneous magnetization axis resulting in a mixing of spins in the spin analyzer (even of internally pure spin states with respect to the instantaneous direction of the local moment).

The photoelectron emission angle is closely related to its internal momentum $\vec{k}$, and, therefore, the reported angular broadening of $\Gamma_{25}'^{\downarrow}$ reflects an internal $\vec{k}$ broadening. The $\vec{k}$ broadening is probably of ferromagnetic origin since it is absent in data taken at $h\nu = 31$ eV, which also in other respects exhibit a different behavior upon heating, as will be shown below.

We have also studied the temperature dependences for $h\nu = 31$ and 21 eV. There is a marked difference from the $h\nu = 60$ eV data of Fig. 3: Instead of a *decrease* in spin-summed intensity near $E_F$ we observe a strong *increase* in total photocurrent upon heating to $T/T_C = 0.85$ for photon energies of 31 and 21 eV. After cooling, the intensity decreases to its initial value. SREDC's taken at $h\nu = 31$ eV (Fig. 4) demonstrate that there is a doubling of $\downarrow$-spin intensity and a comparatively small decrease in $\uparrow$-spin intensity, causing the gain in total intensity. This asymmetry excludes an interpolation of the $\uparrow$- and $\downarrow$-spin intensity changes as transverse fluctuations of a local magnetic moment. We have already interpreted the peak in the $T/T_C = 0.3$ $\uparrow$ SREDC (Fig. 4) as due to direct transitions from the $\Delta_5^{\uparrow}$ band, and from its binding energy we deduce that the transition occurs at about 0.7 of the $\Gamma$-$H$ separation (compare with Fig. 1).

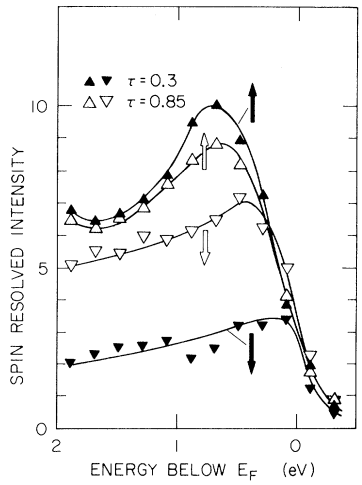


FIG. 4. Spin- and angle-resolved energy-distribution curves of Fe(100) at 31 eV photon energy for $\tau = T/T_C = 0.3$ and 0.85 (unsmoothed data).

Because of the cutoff at $E_F$, the corresponding $\downarrow$ emission from $\Delta_5^{\downarrow}$ does not occur. The strong $\downarrow$-intensity increase upon heating (Fig. 4) suggests therefore that $\Delta_5^{\downarrow}$-symmetry initial states appear at $E_F$ at elevated temperatures near this $k$ vector, making possible efficient direct $\downarrow$-spin phototransitions. This has indeed been predicted by the disordered-local-moment picture to occur as a result of a Stoner-type decrease of the exchange splitting with temperature for $k$ vectors near $H$. The majority-spin phototransition does not suffer from the cutoff at $E_F$, explaining its weaker dependence on temperature.

The new data might serve for testing finite-temperature theories of photoemission from ferromagnets. They have to explain the stationary character of $\Gamma_{25}'^{\uparrow}$, the strong broadening of $\Gamma_{25}'^{\downarrow}$ in both $\vec{k}$ and energy, and the apparent $\Delta_5^{\downarrow}$ band shift for large $k$ vectors. Recent first-principles calculations[5,15] based on the disordered-local-moment picture for Fe are in qualitative agreement with the data.[16]

We note finally that the suggested band shifts could be tested also by the new technique of spin-polarized inverse photoemission[17] since the bands are partially unoccupied in the right half of the Brillouin zone.

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