Wave-Vector–Dependent Temperature Behavior of Empty Bands in Ferromagnetic Iron

J. Kirschner
Institut für Grenzflächenforschung und Vakuumphysik, Kernforschungsanlage Jülich GmbH, D-5170 Jülich, West Germany

and

M. Glöbl, V. Dose, and H. Scheidt
Physikalisches Institut der Universität Würzburg, D-8700 Würzburg, West Germany
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We report on the experimental study of the behavior of unoccupied exchange-split bands in ferromagnets on approaching the Curie temperature. Using spin-polarized isochromat spectroscopy, we observe minority and majority states near the H point in bcc Fe to merge together with increasing temperature, providing evidence for a "collapsing" band state. The flat minority band, generating most of the empty density of states, is sampled at two other points in k space by use of different crystal surfaces. At these locations the position of the peak remains unchanged, indicating the existence of "noncollapsing" band states.

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The electronic structure of itinerant-electron ferromagnets at elevated temperatures currently receives much theoretical and experimental attention. Of particular interest is the temperature dependence of the exchange splitting of energy bands below and above the Fermi level. Recent results of spin-polarized photoemission from Ni showed the energetic splitting between majority and minority peaks from an occupied band near the X point to decrease with increasing temperature. This behavior is consistent with the classical Stoner model, while the increased broadening found on approach to the Curie temperature was not expected from this model.[1] In modern theories[2–7] there appears to be general agreement about the existence of local magnetic moments at temperatures well above the Curie temperature T_C. There is disagreement, however, about the nature of the magnetic structure. While in the "disordered local moment" model[4–7] the moments at different lattice sites are only weakly correlated, in the "fluctuating band theory"[2,3] a strong short-range order over 10–20 Å is assumed. Within the latter approach, the existence of spin waves above T_C in iron could be explained, an experimental result from neutron scattering[8] that is seriously questioned by recent polarized-neutron–scattering experiments.[9] On the other hand, the above findings for Ni (reduction of peak splitting and increased broadening) are shown[10] to be consistent with local band theory even when one assumes a temperature-independent exchange splitting. Within a parabolic band approximation it was also predicted that at other points in the Brillouin zone, peak splittings may persist. Qualitatively similar results have been predicted by the disordered-local-moment theory for the paramagnetic state of iron.[4] In particular, the spin-split empty bands at H_{25}' were predicted to collapse above T_C, i.e., to merge into one single peak. The modifications to the electronic structure of itinerant-electron ferromagnets upon approaching the Curie temperature therefore seem to depend strongly on the location in k space and the band character.

Itinerant-electron ferromagnetism is largely determined by the holes in the d band. This makes experimental investigations of the spin-split band structure in the empty region particularly important. It has recently been demonstrated that spin-polarized isochromat spectroscopy allows mapping of empty minority and majority bands separately.[11] This same approach is used in the present paper to investigate the temperature behavior of selected band states. We find evidence for a collapsing state near the H point of bcc Fe, as well as temperature-independent exchange splittings for the flat band(s) extending over most of the Brillouin zone. As this band provides most of the empty minority density of states, its collapse would lead to major changes in the electronic structure of Fe above the Curie point. Because of the importance of this band, its behavior was studied at two different locations in k space with use of two different crystal surfaces [Fe(110) and (100)], with very similar results.

The experimental setup is the same as described previously.[11] In brief, longitudinally polarized electrons from a GaAsP photocathode hit the single-domain sample at variable angle of incidence from 0° to 85° with respect to the surface normal. The plane of incidence contains the magnetization vector and coincides with a mirror plane of the Fe crys-

tal, thus suppressing spin-orbit effects and their temperature dependence.[12] The bremsstrahlung photons at a particular energy (9.7 eV) are counted by an energy-selective Geiger-Müller counter while the primary energy is slowly increased. The electron spin polarization is reversed with a 2-Hz cycle, interlaced with heating-current and remagnetizing-current pulses. After correction for the effective polarization of the primary beam, the spin-resolved isochromats in Figs. 1 and 2 are obtained for various temperatures between $0.29T_C$ and $0.86T_C$. Extensive cleaning procedures[13] had to be applied to the iron crystals to prevent impurity segregation at elevated temperature.

At room temperature we measured for Fe(100) the dispersion of minority and majority bands with the parallel momentum $\vec{k}$ along the Γ-N direction. These results will be presented in detail elsewhere. Qualitatively, we obtained the same results as on Fe(110): The flat minority band at an average energy of ~ 1.6 eV above E_F agrees quite well with the theoretical data of Wang and Callaway.[14] The empty majority states at large $\vec{k}$ were found to be somewhat closer to the Fermi level than predicted. For the investigation of the temperature dependence of the flat minority band with the (100) surface we chose an angle of incidence of $\theta = 75°$, which brings us to a point about halfway between P and N but not exactly on the D line [see Fig. 1(c)]. The spin-resolved isochromat spectra are shown in Fig. 1(a) for increasing temperature. We clearly see the minority peak at ~ 1.6 eV above the Fermi level to stay in place within ± 0.1 eV, while the polarization decreases gradually. Within the experimental accuracy there is no energy shift to be observed. Also, we cannot find a corresponding majority peak moving up from below the Fermi level, which would be the case if this band collapsed on approaching T_C. It cannot be excluded that the spectral function of this band state develops side peaks[15] near T_C, the experimental resolution being limited to about 0.8 eV full width at half maximum. The results for the (110) face, at $\theta = 60°$, are shown in Fig. 1(b). This geometry corresponds to a point in $\vec{k}$ space near the P-H line in the vicinity of P [see Fig. 1(c)], and the same flat band of minority character is probed. Again, we find a gradual decrease of the polarization but no energetic shift. There seems to be a slight change in the peak shape caused by an intensity increase near the Fermi energy and ~ 0.5 eV above. With due care this finding could be interpreted as an indication for a "central peak" structure predicted by local band theory.[15] The majority counterpart of this band has been observed in conventional photoemission[16] to lie about 0.5 eV below the Fermi energy at P. The exchange splitting at room temperature then amounts to 2.1 ± 0.2 eV.[17] An energetic shift of the occupied majority band upon heating has not been reported.[16]

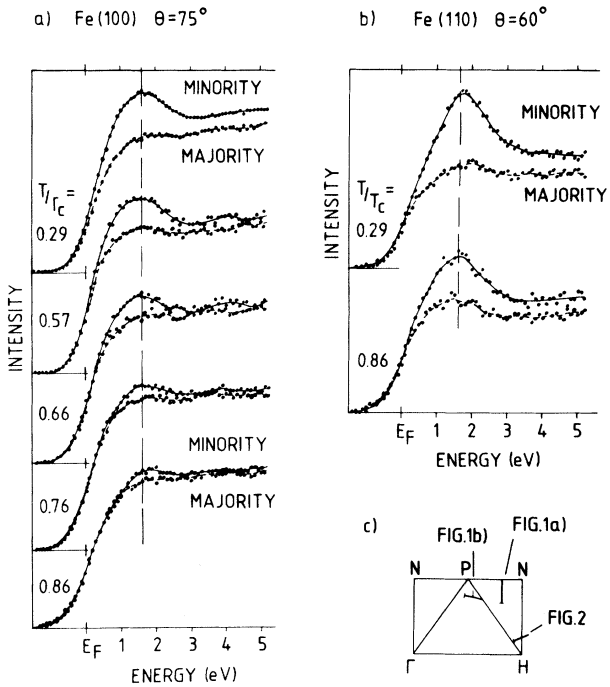

FIG. 1. (a) Spin-resolved inverse photoemission spectra from Fe(100) at angle of incidence $\theta = 75°$ for various reduced temperatures T/T_C. While the difference of the spin-resolved curves decreases with increasing temperature, the energetic position of the maximum in the minority curve remains unchanged (noncollapsing band state). (b) Same as in (a) but for the Fe(110) face at $\theta = 60°$ for room temperature and near the Curie temperature. (c) The path traveled in the Brillouin zone during the isochromat energy scan from 0 to ~ 5 eV above the Fermi energy, calculated with the same parameters as used in Ref. 11. The short bar indicates the starting point.

Quite different behavior is observed near the H point. In the ground state at $T = 0$ the flat minority band is moved slightly away from E_F while its majority counterpart rises somewhat above the Fermi level.[14] Experimentally, this point is reached at $\theta = 0°$, which however, precludes polarization analysis, as for longitudinally polarized electrons the polarization and magnetization vectors are orthogonal.[18] The intensity spectrum [see Fig. 2(a), $\theta = 0°$] shows two peaks at room temperature. By going slightly off normal, the peak near E_F is identified as being of majority type [Fig. 2(a), $\theta = 15°$]. The exchange splitting is found to be

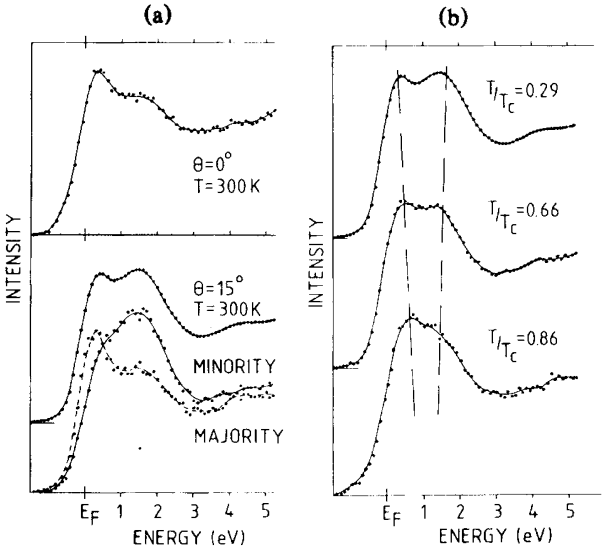

FIG. 2. (a) Total intensity spectra for Fe(100) at $\theta = 0°$ and $\theta = 15°$, taken at room temperature. The spin character of the two-peak structure is identified by the spin-resolved intensity spectra at $\theta = 15°$. For the path in $\vec{k}$ space see Fig. 1(c). (b) Intensity spectra at $\theta = 15°$ for different reduced temperatures T/T_C. Note the merging of the two peaks into one when approaching T_C, indicating a collapsing band state near the H point of the Brillouin zone.

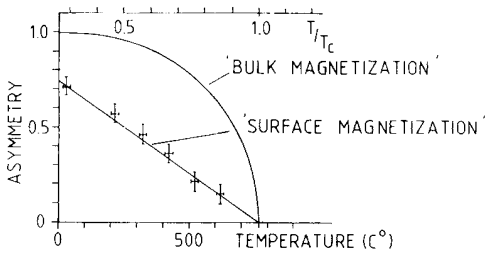

FIG. 3. Intensity asymmetry of the minority and majority intensities in Fig. 1(a) at fixed energy (1.6 eV above E_F) for increasing temperature. Within the experimental error the asymmetry is well fitted by a straight line, taken as being roughly representative of the surface magnetic behavior. For comparison a "bulk" magnetization curve is included.

1.6 ± 0.2 eV at room temperature, somewhat smaller than in theory (2.1 eV).[14] The temperature dependence of the isochromat spectra at $\theta = 15°$ is shown in Fig. 2(b). The majority and minority peaks apparently merge into one single peak located about midway between the room-temperature peaks, indicating a "collapsing" band state at this point in $\vec{k}$ space.[19] These results are in line with a prediction of the disordered-local-moment theory[4]: In the paramagnetic state the Bloch spectral function at H_{25}' consists of a single asymmetric peak which, when folded with our experimental resolution, would have a width of roughly 1.5 eV. We consider our results at least a semiquantitative confirmation of this prediction.

A few remarks concerning the surface nature of the present experiments shall be made. Our interpretation is based entirely on the bulk band structure. This is probably justified because realistic calculations (including the surface) of spin-polarized isochromat spectroscopy[20] showed this approach to be valid in most cases as far as energetic positions of spin-resolved intensities are concerned. The magnetization measured in the experiment, however, is not the bulk magnetization. Because of the magnetic coherence length increasing towards T_C, bulk and surface magnetizations will be sampled to different degrees, depending on temperature. A rough estimate of this effect may be obtained from the asymmetry of the noncollapsing band in Fig. 1(a). Within experimental error the asymmetry is proportional to $T_C - T$, as shown in Fig. 3. Assuming the asymmetry to be proportional to the magnetization, we find a typical surface magnetism behavior, with the magnetization decaying much faster with increasing temperature than in the bulk. This is probably the reason why relatively strong effects, normally expected for $T/T_C > 0.9$, here occur at much lower temperature. The surface behavior is not too surprising in view of a depth of information of about 2 to 3 monolayers[21] at large angle of incidence. Near normal incidence it is roughly a factor of 4 larger.

In conclusion, we find that whether an empty band state collapses on approaching the Curie temperature or not depends strongly on the $\vec{k}$ point selected in the experiment. We note that states on a flat band with small dispersion in the vicinity of the $\vec{k}$ point chosen appear not to collapse. By contrast, when bands disperse strongly in the vicinity of the observed band state, it tends to collapse towards T_C.

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[18]The price to be paid for high intensity and suppression of spin-orbit effects is an angle-dependent effective polarization (see also Ref. 13). This handicap at small angles of incidence can be compensated by extended measurement times at room temperature. At high temperatures the measurement time is limited by impurity segregation and the intensity is reduced by the duty cycle of pulsed heating and by the Debye-Waller factor. These factors together precluded the measurement of full spin-resolved spectra at elevated temperature. The spin character of the respective states was, however, verified by spin-resolved measurements at several fixed energies.

[19]The influence of the thermal lattice expansion is considered negligible for the following reason: Over the present temperature range the iron lattice expands linearly by about 0.7%. A theoretical band-structure calculation near the H point with a 1% increased lattice constant yielded an energy shift by 0.1 to 0.2 eV towards the Fermi level for the minority *and* the majority bands (R. Feder and A. Rodriquez, private communication). A resultant shift by ~ 0.1 eV is below the resolution limit of the present experiment.

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