Exchange-Split Empty Energy Bands of Fe(110)

H. Scheidt, M. Glöbl, and V. Dose
Physikalisches Institut der Universität, D-8700 Würzburg, Germany

and

J. Kirschner
Institut für Grenzflächen und Vakuumphysik der Kernforschungsanlage, D-5170 Jülich, Germany

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Spin- and momentum-resolved bremsstrahlung isochromat spectra from Fe(110) at $\hbar\omega = 9.7$ eV are presented, a combination which allows for the first time the determination of experimental final-state dispersions $E(k_{\parallel})$ for direct interband transitions into majority and minority final bands separately. Data are in good agreement with relevant theoretical $E(k_{\parallel})$ values from bulk band-structure calculations as far as minority final states are concerned. Theoretical predictions for the majority states exhibit a stronger dispersion than observed experimentally.

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Considerable progress towards an understanding of the ferromagnetism of the 3*d* metals has recently been achieved through the development of the fluctuating local band theory.[1-4] In this theory, the magnetic phase transition is essentially determined by time- and space-dependent fluctuations in the direction of the local magnetization. Though the improvement of the predictive power of this approach is impressive compared to the Stoner-Wohlfarth theory, further quantitative development appears to require a precise experimental knowledge of the band structure of these materials.[5] Occupied and unoccupied electronic states are equally important in this context. In the case of Ni, it has been argued that the holes in the *d* band are the "active ingredient" of the magnetism.[6] It is also essential to resolve the spin quantum number in the experiment. While the sign of the polarization allows a straightforward experimental identification of majority and minority bands, the magnitude of the average spin polarization within a band may serve as a critical test of the predictions of the fluctuating band theory. For occupied states, momentum- and spin-resolved photoemission has been shown to be feasible[7] and was recently applied to Fe-(100).[8,9] The most promising technique for the investigation of the empty electronic states appears to be spin-polarized inverse photoemission. In particular, the most interesting region between the Fermi and vacuum levels is hardly accessible otherwise. A first step in this direction has been reported by Unguris *et al.*[10] An empty minority band of Ni near the *X* point of the Brillouin zone was identified. The second and crucial step is to measure both empty minority and majority bands individually, and to map out their dispersion separately. This is the subject of the present paper. We present spin- and angle-resolved inverse photoemission spectra from Fe(110) for angles of electron incidence between normal and glancing which allow the identification of two minority bands and one majority band from structures not discernible in the spin-averaged data.

A sketch of our experimental arrangement is shown in Fig. 1. Spin-polarized electrons are obtained from a negative-electron-affinity GaAsP photoemitter.[11] The polarization is parallel or antiparallel to the electron momentum depending on the helicity of the circularly polarized He-Ne-laser radiation incident on the photocathode. With longitudinal electron polarization, complications

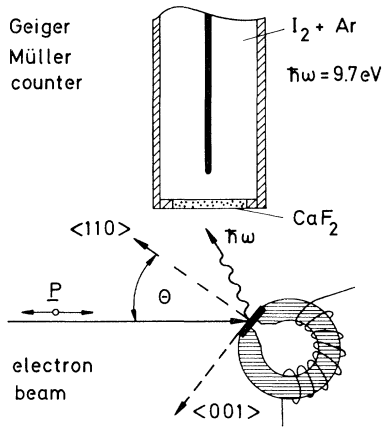

FIG. 1. Schematic of the apparatus for momentum-resolved, spin-polarized bremsstrahlung isochromat spectroscopy.

due to spin-orbit coupling vanish in the present geometry. Moreover, it allows large currents which were of the order of up to 20 μA at 10 eV in the present case. The degree of polarization was close to 35%. The electrons impinge on the sample at polar angles $0 \leq \theta \leq 80°$ in the $\langle 001 \rangle$ azimuth. This is also the direction of sample magnetization which was effected by a small C-shaped electromagnet.

Light emitted by the sample is detected by an energy-selective Geiger-Müller counter which offers a band pass of 9.7 ± 0.35 eV, a high sensitivity of the order of 10^{-2} counts per photon and, if operated properly, a very low background of about 5–10 counts per minute.[12] This is negligible in comparison to our signal level of 10^3 s^{-1}. Sample preparation included argon-ion etching and annealing. Sample cleanliness was monitored by (cylindrical-mirror analyzer) Auger analysis. The annealing temperature was critical and held below 580 °C in order to avoid sulfur segregation. The observed spin-dependent asymmetry A in the photon production is

$$A = (N_+ - N_-)/(N_- + N_+), \quad (1)$$

where $N_\pm$ indicate the photon emission per unit incident charge. Let $n_\pm$ be the photon production for a hypothetical 100%-spin-polarized beam. We then have that

$$n_\pm = \tfrac{1}{2}(1 \pm A/P_0 \sin\theta)(N_- + N_+), \quad (2)$$

where P_0 denotes the actual spin polarization.

Figure 2 shows a sample of our isochromat spectra for various angles of incidence θ. The upper curve in each panel represents the spin-averaged data, $\frac{1}{2}(n_- + n_+)$. All spin-averaged data have been normalized to the same peak emission. A normal-incidence spectrum has been added to the $\theta = 20°$ panel. The spin-averaged data show two separated emission features for $\theta \leq 20°$ but exhibit only one rather broad emission maximum for $\theta \geq 25°$. The rather similar shape of the spin-averaged data for $\theta \geq 25°$ emphasizes the necessity for spin resolution in order to derive more detailed information. Spin-resolved isochromats are plotted as the lower full dots in Fig. 2 for transitions into empty majority-spin bands and as open circles for transitions into minority states. The statistical uncertainty is indicated by the error bars. Full or dashed lines simply connect consecutive data points to guide the eye. n_+ spectra show a single well-resolved maximum which disperses with increasing polar angle to higher energies and gradually fades out for $\theta \geq 60°$. The

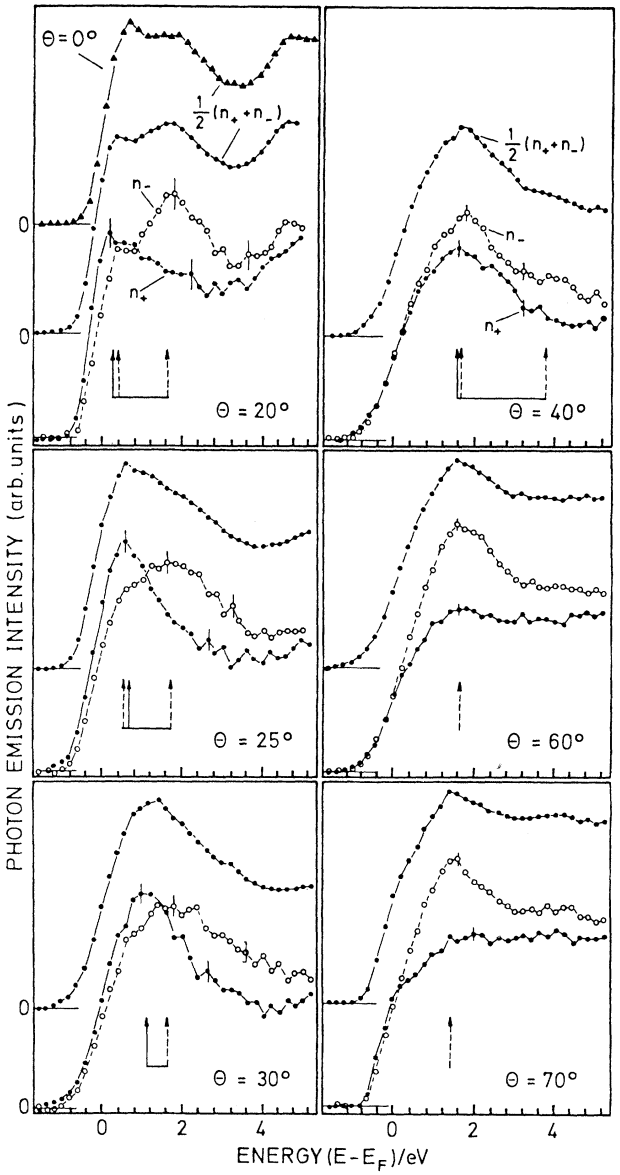

FIG. 2. Spin-averaged and spin-resolved isochromats at various angles of incidence. Emission from transitions into empty minority states is marked by open circles. Upper full dots represent spin-averaged data, while the lower full dots indicate emission from transitions into majority final states.

energetic position of this emission maximum is indicated by solid arrows. n_- spectra exhibit two emission maxima for $\theta \leq 25°$. For $\theta = 30°$, only one emission feature remains discernible. However, a broad shoulder in the 2–4 eV region suggests a further contribution at higher energies which apparently splits off in the $\theta = 40°$ spectrum as a weak feature at about 3.8 eV. The energetic positions of n_- emission features are indicated

by dashed arrows. Peak positions as a function of $k_{\parallel}$, the component of electron momentum parallel to the sample surface, are summarized as full dots in Fig. 3(b) and represent the first experimental band dispersions $E(k_{\parallel})$ for minority and majority states separately. One majority final band with rapid dispersion and two minority final bands are observed, one of which shows practically no dispersion. An indication of a third band is found. Data points from spectra for polar angles other than in Fig. 2 have also been included.

We shall now demonstrate that all observed emission features can be interpreted in terms of radiative transitions between unoccupied bulk energy bands. To this end, we first give an estimate of the range of bulk k values which is covered by the present sample of isochromats in the ΓHPN mirror plane of the bcc Brillouin zone. The irreducible part of this mirror plane in k space is shown in Fig. 3(a). In analogy to common practice in angle-resolved photoemission, we assume a free-electron dispersion for the initial state. The corresponding constant-initial-energy curves in k space are circles around Γ. Such circles reduced to the first Brillouin zone by the lattice vector $(2\pi/a)(\bar{1}\bar{1}0)$ are shown in Fig. 3(a). The circles have been calculated using an inner potential of $V_i = 12$ eV as determined by low-energy electron diffraction investigations[13] and a work function of $e\varphi = 4.5$ eV. Energy labels refer to the final-state energy $E_f = E_i - 9.7$ eV. The dashed curves intersecting the circles describe the paths in k space when for a given polar angle the energy is scanned in the isochromat measurement.

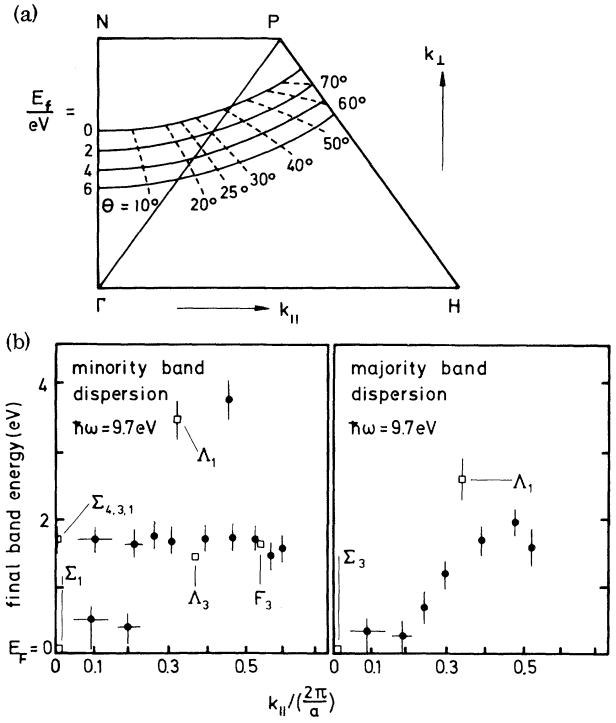

FIG. 3. (a) The irreducible section of the ΓHPN mirror plane of the bcc Brillouin zone. Circles are free-electron constant-energy curves. Dashed lines are the paths in k space for the isochromat energy variation at given polar angles. Full dots in (b) show the dispersion of emission features marked by arrows in Fig. 2. Rectangles indicate theoretical $E(k_{\parallel})$ values for bulk direct transitions deduced from the bulk band structure of Fig. 4.

In order to compare our experimental final-state energies to band-structure calculations[14] which are available on high-symmetry lines in k space, we shall now consider possible radiative transitions with $\hbar\omega = 9.7$ eV between calculated initial and final states on the H-P, Γ-P, and Γ-N lines. Those transitions are indicated in Fig. 4 by full and dashed arrows for majority and minority final states, respectively. On H-F-P, only one transition is possible within the relevant final energy range at $E_f = 1.6$ eV. Many final Σ states are available on the Γ-N line: a Σ_1 minority and a Σ_3 majority state near the Fermi level and three minority states of Σ_1, Σ_3, and Σ_4 symmetry at about 1.7 eV indicated by only one dashed arrow. On the Γ-P high-symmetry line, transitions are possible into one Λ_1 majority final state and its adjoining exchange-split minority

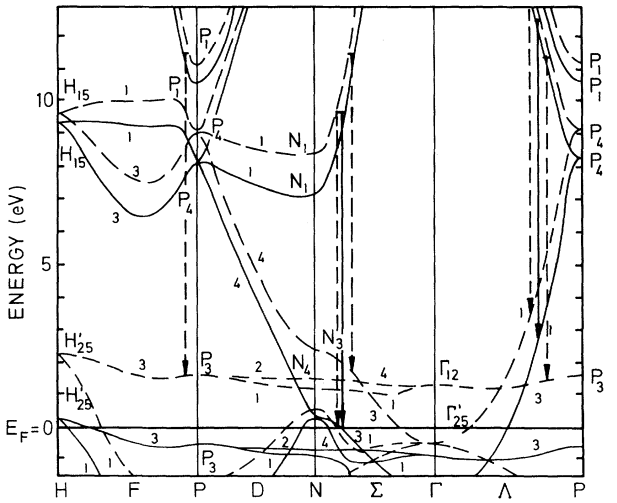

FIG. 4. Ferromagnetic band structure of iron (Ref. 14). Dashed and full arrows indicate possible bulk direct transitions for $\hbar\omega = 9.7$ eV.

states at 2.6 and 3.5 eV, respectively. A third arrow indicates a transition into a Λ_3 minority state at 1.45 eV. The arrow position on the abscissa completely determines the reduced bulk k vector of the transitions concerned. This leads, together with the final band energies, to theoretical $E(k_\parallel)$ values which are plotted as rectangles in Fig. 3(b) and marked by the corresponding final-state symmetry. Whereas the agreement between theory and experiment is good for transitions into final minority states, the theoretical dispersion of the majority band deviates significantly to higher energies even if one takes a large error bar due to rapid band dispersion into account.

We want finally to comment briefly on the residual polarization above 4 eV in all spin-resolved spectra. An apparatus asymmetry has been excluded by reversing the sample magnetization. It is well known, however, that isochromat spectra also contain contributions from electron-hole pair production prior to the radiative transition.[15] These secondary contributions become progressively important as the final-state energy moves away from the Fermi level and depend strongly on the density of empty states. The higher minority radiative emission should then be a consequence of the considerable asymmetry due to a much larger density of empty minority states as compared to empty majority states.

In summary, we have presented for the first time spin-resolved empty-state dispersion for an itinerant ferromagnet. Theoretical $E(k_\parallel)$ values for bulk direct transitions in minority states are in good agreement with experimental data. The theoretical results for majority final states lead to a stronger dispersion than observed experimentally. Moreover, the results presented here demonstrate that the technique used is a promising tool to attack the remaining problems in ferromagnetism, such as the experimental identification of empty magnetic surface states or the temperature dependence of exchange-split bands.

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