Theoretical limitations to the determination of bandwidth and electron mass renormalization: the case of ferromagnetic iron

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Abstract. Recent experimental advances have allowed electronic band structures to be investigated by angle-resolved photoemission in considerably more detail. A recent study of ferromagnetic iron finds the occupied bandwidth, for the two shallow bands observed, reduced by $\sim 30\%$ as compared to the calculated ground state, rendering bcc iron comparable with the strongly correlated transition metal Ni. Fermi velocities were reported to deviate from the ground state and these deviations have been assigned entirely to electron correlation. We show that spin–orbit splitting, final-state transitions and final-state broadening significantly change the band dispersion as measured by a modern energy analyzer, and a simple model that accounts for their effects is introduced. Applying our model, we find for the occupied bandwidth a narrowing of the order of only $10\%$ in agreement with the literature. Substantial renormalization of the Fermi velocities is confirmed but a significantly smaller fraction of it is attributed to correlation effects, namely many-electron interactions.

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1. Introduction

Angle-resolved photoemission spectroscopy (ARPES) is the major experimental method to
investigate the electronic structure of solids (Hüfner 1995, Kevan 1992). It directly probes the
electron-wave-vector dependence of occupied electronic states. The photoemission spectrum
is modeled by a spectral function $A(k, E)$ with electron wave vector $k$ and binding energy $E$
which includes electron–electron interactions by a complex self-energy (Braun 1996, Kevan
1992, Pendry 1976). Band structures of metals are often successfully calculated by density-
fuctional theory (DFT) without self-energy corrections. For certain solids such as the noble
metal Cu, the one-electron band-structure picture of DFT has been found to agree very well
with the $E(k)$ dispersion obtained by ARPES (Thiry 1981, Thiry et al 1979). However, for
its neighbor in the periodic system, the 3d transition metal Ni, the strong effects of electron
correlation on core-level (Hüfner and Wertheim 1975) and valence–band spectra (Guillot
et al 1977) have been established. The occupied 3d bandwidth is reduced by $\sim 30\%$ with respect to
density-functional calculations, and the ferromagnetic exchange splitting by $\sim 50\%$ (Eberhardt
and Plummer 1980, Himpsel et al 1979). It is believed that the localized character of the
3d orbital leads to enhanced electron correlation and that this causes the deviations from the
band structure picture. The deviation between DFT in the local-density approximation (LDA)
and ARPES can therefore be considered a measure of electron correlation. The enhanced
electron correlation in the 3d orbital is also the cause of ferromagnetic and anti-ferromagnetic
interactions in transition metals. For these reasons, the question posed for the other magnetic
3d transition metals is: how well does the band dispersion calculated for the ground state by the
LDA agree with the one obtained by ARPES? For Cr, Fe, Co and Ni, detailed band dispersions
have been measured and, with the exception of Ni, good agreement in binding energies within
10% has been found (Rader and Gudat 1999). For Fe, in particular, an evaluation of the energy
range from 5 eV below the Fermi energy ($E_F$) to 2 eV above $E_F$ gave a compression of the
bands by 10% (Santoni and Himpsel 1991). Thus, solely based on bandwidth arguments, Ni is
strongly correlated, whereas Fe is not. On the other hand, the photoemission peaks show a large
linear broadening ($\sim 60\%$ of the binding energy) in Fe (Santoni and Himpsel 1991). The strong
broadening was predicted to be accompanied by a strong loss in spectral weight (Katsnelson
and Lichtenstein 1999) and, for Co, this effect was confirmed and studied in detail. The majority-
spin bands lose intensity for binding energies larger than 2 eV (Monastra et al 2002) and
are difficult to detect even in spin-resolved spectroscopy (Alkemper et al 1994). Many-body calculations identified this as a correlation effect with increased Coulomb correlation energy, $U$, leading to a stronger damping (Monastra et al 2002). An important sign of strong electron correlation in Ni is the appearance of photoemission satellites in the core-level (Hüfner and Wertheim 1975) and valence-band spectra (Guillot et al 1977, Höchst et al 1977). For Fe, a satellite at 7 eV was predicted by a many-body calculation for a relatively high value of $U$ of 2.3 eV but is not observed in experiment (Grechnev et al 2007). In resonant photoemission at the Fe L-edge, a satellite was observed at a binding energy of $\sim$3 eV (Hüfner et al 2000) but it has not yet been established why, unlike in Ni, it is not observed at low photon energies at or below the M-edge.

Recent experimental advances in ARPES have allowed energy resolutions of a few meV and angle resolutions around 0.1° to be obtained. This has led to renewed interest in the question of electron correlation effects on the $E(k)$ band dispersion of the magnetic transition metals. The high angle and thus $k_\parallel$ resolution (where $k_\parallel$ denotes the projection of $k$ onto the surface plane of the sample) and especially the simultaneous detection of a range of $k_\parallel$ values allow for precise measurement of the electron binding energy, velocity and effective mass. In a study of surface states of Fe(110), a considerable renormalization of the electron velocity, i.e. a deviation from the LDA value, with a kink on the energy scale of spin-wave excitations ($\sim$125 meV), has been observed (Schäfer et al 2004). The above seminal work on the near-surface electronic structure was recently followed by studies on bulk states of Fe, and indications of a strong electron correlation were observed there as well (Schäfer et al 2005, Cui et al 2006, 2007). Bandwidth reductions of 27% for the minority-spin states of Fe, and of 33% for majority spin were obtained (Schäfer et al 2005). Because of simultaneous measurement of a $k_\parallel$ range, modern energy analyzers are particularly suited for measuring the Fermi velocity $v_F$. In the recent experiment, large mass renormalizations of $v_F$ up to a factor of 3.5 or more were obtained (Schäfer et al 2005). These are assigned to strong electron correlation (Schäfer et al 2005). We will show that other factors contribute to the observed deviations, and inclusion of various aspects of the photoemission process and consideration of correlation effects, via the imaginary component of self-energy, in the theoretical description lead to an improved modeling of the experimentally observed spectra and to lower bandwidth and mass renormalization values.

The present paper reviews the determination of bandwidth and mass renormalization including all relevant effects in the LDA calculation and the photoemission process. We show how neglect of each of these effects results in a different and hence model-dependent renormalization value. One-step photoemission calculations of Fe (Redinger et al 1988) could also be used to address these questions but, to our knowledge, this has not been reported as yet. The advantage of our treatment is that of conceptual and computational simplicity. We examine the limitations of the recently published analysis (Schäfer et al 2005) and demonstrate that the use of a more complete model, including among others final-state photoemission effects, leads to a drastic decrease in the bandwidth renormalization and a significant decrease in the mass renormalization observed. All published values are re-examined and the new mass-renormalization values show no relation between Fermi-sheet size and high electron correlation, or any relation to the proximity of smaller sheets of opposite spin, as speculated previously (Schäfer et al 2005).
2. Previous bandwidth and mass renormalization studies

A study of Fe was undertaken (Turner et al 1984) in which ARPES measurements were compared with three-step photoemission calculations. The theoretical model for the initial states was the DFT in the local spin density approximation (LSDA) published for Fe previously (Callaway and Wang 1977). A free electron final state (FEFS) approximation was used to determine the expected transitions and a correlation potential was used in the calculations to obtain agreement between the theoretical and the experimental band dispersion. Only a small correlation potential, as compared with that from Ni investigations, was required to correctly describe the bands to within experimental error.

ARPES data on Fe with much higher energy and angle resolution than those measured 25 years ago (Turner et al 1984) have been presented in a recent publication (Schäfer et al 2005). Comparison of the higher resolution data, however, is made with the calculated initial state only. Calculation of the initial states was performed using the augmented-plane-wave-plus-local-orbitals (ALW + LO) potentials, and scalar relativistic effects were included but spin–orbit coupling effects were neglected. Fermi vectors and Fermi velocities are derived from the initial states and compared with experimental results, with the Fermi velocity ratio (theoretical velocity over experimental velocity) used to determine the mass renormalization. A second aspect of comparison between theory and experiment employed is the bandwidth renormalization, calculated as the ratio of theoretical over experimental band extrema at critical points. The mass renormalization values obtained (Schäfer et al 2005) lie in the range of 1.5–3.6, with bandwidth renormalization values of 1.4 and 1.5 at the \( \Gamma \) and \( P \) points, respectively. From the obtained renormalization values, a non-uniform scattering mechanism is speculated for the electron correlation with an increase in the scattering caused by proximity to a smaller Fermi-surface sheet of opposite spin. Moreover, larger Fermi surface sheets were found to lead to higher mass renormalization (Schäfer et al 2005).

Recently, a theoretical investigation of the correlation effects in Fe, Ni and Co was performed (Grechnev et al 2007). In this study, LDA calculations were used in conjunction with the dynamical mean field theory (DMFT) formalism, and the resulting electron self-energy, density of states and the spectral density were discussed. The correlation parameters, \( U \) and \( J \), were found to be substantial in Fe and Co (2.3 and 0.9 eV) but smaller than those in Ni (3 and 1 eV). In particular, \( U \) is so large that a correlation satellite at \( \sim 7 \) eV was predicted, which is, however, not observed in experiment. For the binding energies in the range 0 –1 eV, consideration of figures 3, 17 and 18 (Grechnev et al 2007) appears to indicate a real component of the self-energy varying from 0 to 1 eV in Fe. In particular, at the expected critical point energies at \( \Gamma \) (minority) and \( P \) (majority), this corresponds to a bandwidth renormalization of about 50%. This is higher than that observed from comparison between the experiment and initial states alone (Schäfer et al 2005) and is even higher than that previously established for Ni (\( \sim 30\% \)).

3. Spin–orbit splitting of initial states and inclusion of transitions to the final state

The present modeling employs the same APW + LO initial state calculations including scalar relativistic effects as used before (Schäfer et al 2005). These calculations were performed using WIEN2k computer code (Blaha et al 2001). It is expected that the spin–orbit coupling affects the valence bands of Fe, and a spin–orbit splitting of 110 meV has been observed in

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ARPES (Sakisaka et al. 1990). The present calculations use the second variational method to approximate spin–orbit coupling. This accounts for spin–orbit splitting and changes of the band topology since bands of like symmetry do not cross. As these two calculations only consider the initial states, they will be labeled the initial state model (ISM) and the initial state model plus spin–orbit splitting (ISM + SO), respectively. The ISM calculations of Schäfer et al. (2005) and the ISM + SO are shown in figures 1(a) and (b), respectively. Differences are observed in bands II, III and IV where the spin–orbit interactions cause extra hybridization with subtle changes of shape and a separation of the bands III and IV; a comparison of the critical point energies indicates significant differences. Fermi vectors, Fermi velocities and mass renormalization values for the two models are included in table 3 and will be discussed in section 5.

Photoemission experiments probe the transitions from the initial states to a final state above the Fermi energy. Our next step in developing an appropriate model is to include these transitions in the calculation. We will use the FEFS approximation, as was done by Turner et al. (1984). The FEFS approximation is widely used in photoemission experiments and assumes the final state to be of the form

\[ E(k) = \frac{\hbar^2 k^2}{2m} + V_0 \]  

The inner potential, \( V_0 \), was determined from experimental ARPES as 14.8 eV (Schäfer et al. 2005). Transitions from the ISM + SO initial states to the FEFS via the photon energy, \( \hbar \omega \), will be considered in the direct transition model (DTM). Equation (1) indicates that the three-dimensional \( k \)-space shape of a FEFS is spherical with a radius related to the band energy and hence the photon energy of the transition. The allowed transitions are determined as the intersection of the FEFS with the three-dimensional ISM + SO initial state calculations. The DTM results for a photon energy of 139 eV in the [100] direction of (110) BCC Fe are shown in figure 1(c). Due to the inclusion of the final state, the wave-vector component perpendicular to the sample surface, \( k_\perp \), is no longer constant with varying \( k_\parallel \). This means that when comparing figure 1(b) with figure 1(c), at the same value of \( k_\parallel \) where figure 1(b) shows H, in figure 1(c) the deviation of the probed \( k \)-point from H is 0.4 Å \(^{-1} \) or 18% of the distance from \( \Gamma \) to H. This causes significant energy shifts. For example, close to the Fermi energy a significant change in bands II, III and IV occurs at H with similar changes for several other bands at higher binding energies. At \( \Gamma \) there is no appreciable change, indicating that the FEFS at the chosen photon energy of 139 eV intersects the \( \Gamma \) point as indicated previously (Schäfer et al. 2005). Determination of the allowed transitions between the initial and final states does not exhaust the theoretical process of ARPES; in addition, broadening of these transitions occurs. A procedure to include this broadening in the current calculations is presented in section 4.

4. Broadening of the electron transitions

In previous analyses of ARPES data of Fe (Fedorov et al. 2002, Schäfer et al. 2004, 2005, Turner et al. 1984, Yamasaki and Fujiiwara 2003), the effect of broadening has, to our knowledge, not been considered. The broadening has been investigated via a one-step model calculation (Redinger et al. 1988) but was limited to low photon energies and normal emission; in contrast, the current calculations were performed for high photon energies (139 eV) and for a range of emission angles. Note that the effect of broadening processes on the Fermi maps and

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Figure 1. Band structure calculations of ferromagnetic Fe: (a) without spin–orbit splitting and final state transitions; (b) with spin–orbit splitting; (c) with spin–orbit splitting and free electron final state transitions (139 eV photon energy), the Brillouin zone boundaries are shown in grey. (a and b) $E(k)$ and (c) $E(k_{\parallel})$ with $k_{\parallel}$ along [001]. Majority-spin bands are shown in red and minority-spin bands in blue. (b) Spin–orbit splitting changes the band structure throughout the Brillouin zone, whereas (c) inclusion of a free-electron final state reveals that for larger $k_{\parallel}$ values, comparable to the $\Gamma$–H distance, the probed region of $k$-space is very different from the initial-state assumption, leading to dramatic changes of binding energies and Fermi velocities for bands II, III and IV near $E_F$.

band maps is not just to smear out the observed bands but to also allow the bands close to the intersection with the FEFS to be observed. Two distinct broadening processes have been described previously (Strocov 2003): broadening of the initial state caused by the finite hole lifetime, $\tau_h$, and broadening of the final state wavefunction due to inelastic absorption and elastic reflection from the crystal potential. The final state spectral function, $A_{\text{final}}(k_{\perp})$, is characterized (Strocov 2003) by a Lorentzian distribution in $k_{\perp}$ centered on the real component of $k_{\perp}(k_{\perp}^0)$ and with a width, $\delta k_{\perp}$, related to the inelastic electron mean free path, $\lambda$, via the relation $\delta k_{\perp} = 2 \text{Im}(k_{\perp}) = 1/\lambda$. The finite hole lifetime initial state spectral function, $A_{\text{initial}}(k_{\perp})$, is characterized by a Lorentzian distribution in energy centered on the band energy and with a full width, $\delta E$, given by $\delta E = h/\tau_h$. This broadening is a result of correlation and
Figure 2. Broadened-transition-model calculation (yellow) of the band structure with $k_\parallel$ along the $\Gamma$–P direction at 139 eV photon energy. Overlaid in red (majority spin, solid) and blue (minority spin, dashed) are the direct-transition-model calculations (a, b and c) and the initial state calculations (Schäfer et al 2005) (d, e and f); the Brillouin zone boundaries are shown in grey. Insets are magnified views of bands I (b and e) and VI (c and f) indicating the change in gradient over 1 eV (b and e) and 0.4 eV (c and f), more than that expected for electron–magnon and electron–phonon interactions.

is generally attributed to the imaginary component of the self-energy (Grechnev et al 2007, Monastra et al 2002) and, in conjunction with the real component of the self-energy, has in particular been shown to increase the agreement between experiment and theory in Co (Grechnev et al 2007, Monastra et al 2002). The photoemission current, $I(E_{\text{final}}, E_{\text{initial}})$, is then of the following form (Strocov 2003):

$$I(E_{\text{final}}, E_{\text{initial}}) \propto \int |T_{\text{final}}|^2 |M_{fi}(k_\perp)|^2 \left( \frac{\delta k_\perp}{(k_\perp - k_\perp^0)^2 + (\frac{\delta k_\perp}{2})^2} \right) \times \left( \frac{\delta E}{(E_{\text{final}} - E_{\text{initial}}(k_\perp))^2 + (\frac{\delta E}{2})^2} \right) dk_\perp,$$

(2)

where $|T_{\text{final}}|$ is the final state surface transmission factor and $|M_{fi}(k_\perp)|$ is the photoexcitation matrix element. In the calculation of the spectral function below, these two terms are set to unity.

The values for $\delta k_\perp$ and $\delta E$ have been determined by comparison with experiment. Figure 2 shows an example calculation that compares allowed transitions, with and without broadening, and the initial state calculations along the [1–11] direction of (110) BCC Fe (figure 10; Schäfer et al 2005). The diffuse intensity (yellow) in figure 2 is a calculation according to our broadened transition model (BTM). It is in excellent agreement with the experimental data. From such comparisons, $\delta k_\perp$ was found to be 0.3 Å$^{-1}$. This result is consistent with the electron inelastic mean free path $\lambda$ (Tanuma et al 1991, Werner et al 2000). From a linear interpolation between the values for 85 and 150 eV (Tanuma et al 1991), we obtain $\lambda \approx 5$ Å for Fe or $\delta k_\perp \approx 0.2$ Å$^{-1}$. Momentum broadening in the band structure regime is frequently represented as a fraction of the Brillouin zone width $k_{\perp}^{\text{BZ}}$ and values lying in the range of 10% (Paggel et al 2000) to
Table 1. Fermi vectors and Fermi velocities calculated using a constant energy broadening ($\delta E = 0.4$ eV) and energy broadening reduced via the relation $\delta E = 3.47 E^2$ for binding energies $E_{\text{initial}}(k_{\perp}) < 0.34$ eV. The difference is negligible, indicating that the momentum broadening provides a significant contribution.

| Fermi sheet | BTM (var. $E$ broad) $k_F$ ($\text{Å}^{-1}$) | Slope (eV/$\text{Å}$) | BTM (const. $E$ broad) $k_F$ ($\text{Å}^{-1}$) | Slope (eV/$\text{Å}$) |
|-------------|----------------------------------|-------------------|----------------------------------|-------------------|
| VI          | 0.27                             | 1.33              | 0.28                             | 1.33              |
| I           | 0.97                             | 3.14              | 0.97                             | 3.11              |
| VI          | 0.32                             | 0.79              | 0.34                             | 0.81              |
| I           | 1.16                             | 1.31              | 1.16                             | 1.23              |
| VI          | 0.39                             | 0.81              | 0.41                             | 0.9               |
| V           | 1.03                             | 1.16              | 1.08                             | 1.15              |
| I           | 0.95                             | 1.41              | 0.96                             | 1.43              |
| II          | 1.79                             | 0.81              | 1.72                             | 0.83              |
| V           | 0.69                             | 1.97              | 0.69                             | 1.47              |

We find that the $\delta k_{\perp}$ broadening dominates over the effect of $\delta E$. Fermi-liquid theory predicts a quadratic dependence of $\delta E$ on the binding energy $E_B$ but only a linear broadening with $\delta E \approx 0.6 \, |E_B|$ was extracted from direct and inverse photoemission data of Fe along the surface normal (Santoni and Himpsel 1991). Most recently, high-resolution low-temperature (10 K) data of bulk states of Fe (110) allow the extraction of a quadratic dependence in the vicinity of $E_F$ (Cui et al 2006, 2007). The current model employs a quadratic dependence of $\delta E$ on $E_B$ close to the Fermi energy with a smooth transition to a constant $\delta E$ at higher binding energy. We use the fit applied to the two majority- and minority-spin bulk bands (Cui et al 2006, 2007), which is described by $\delta E = 3.47 \, E_B^2$ (eV)$^{-1}$. In order to stay within the known broadening (Santoni and Himpsel 1991), we use the quadratic behavior down to $E_B = 0.34$ eV and a constant value (0.4 eV) in the energy range $0.34$ eV $< E < 1$ eV. This behavior is similar to the imaginary component of the self-energy in Fe calculated using DMFT (Grechnev et al 2007). Table 1 presents the calculated BTM Fermi vectors and Fermi velocities obtained using this parameterization for $\delta k_{\perp}$ and $\delta E$ (‘BTM var. $E$ broad’ in table 1). Our results do not depend greatly on the choice of $\delta E = \delta E(E_B)$. In fact, even the choice of a constant $\delta E = 0.4$ eV up to $E_F$ does not change the results for the mass renormalization appreciably (‘BTM const. $E$ broad’ in table 1). Overlaid on figure 2 are the DTM calculated bands for majority spin (red solid lines) and minority spin (blue dashed lines). A comparison of the DTM and the BTM shows how strongly the broadening affects the band dispersion. For the two bands that cross the Fermi surface (bands I and VI) there are significant differences in the Fermi wave vectors and velocities, which affect the bandwidth and mass renormalization values.

5. Experimental comparison: bandwidth and mass renormalization

Bandwidth and mass renormalization values for all the transition models discussed above will be compared to show the effect of the choice of the model on the data analysis. Only two
Table 2. Critical point energies (in eV), bandwidth renormalization and bandwidth reduction values for different theoretical models.

| Position | Experiment (Schäfer et al 2005) | ISM | ISM + SO | DTM | BTM |
|----------|----------------------------------|-----|---------|-----|-----|
|          | error margin ±20%                |     |         |     |     |
| Critical point energy (eV) | Γ | 0.19 | 0.26 | 0.22 | 0.22 | 0.192 |
|          | P | 0.57 | 0.85 | 0.75 | 0.72 | 0.65 |
| Bandwidth renormalization (bandwidth reduction) | Γ | n/a | 1.4 | 1.16 | 1.16 | 1.01 |
|          | P | n/a | 1.5 | 1.32 | 1.26 | 1.14 |

Figure 3. Plot of bandwidth renormalization (a) and reduction (b) as a function of model complexity: initial state model (ISM); initial state model plus spin–orbit interactions (ISM + SO), direct transition model (DTM) and broadened transition model (BTM).

Experimental occupied band widths are observed in the experimental data: the minority-spin band VI at Γ and the majority-spin bands III–II at P. The values of critical point energy, bandwidth renormalization (ratio of the theoretical to the experimental critical point energy) and bandwidth reduction (percentage difference in critical energy) are shown in table 2. Figure 3 demonstrates that the bandwidth renormalization and bandwidth reduction are significantly reduced as the complexity of the model is increased. The BTM gives bandwidth renormalization (bandwidth reduction) values of 1.01 (1%) at Γ and 1.14 (12%) at P, significantly less than in the previous ISM analysis (Schäfer et al 2005). This is in contradiction to the significantly high values (∼50%) obtained via LDA + DMFT calculations (Grechnev et al 2007). The calculated values are also significantly larger than those determined by comparison of experimental data with the initial states alone (Schäfer et al 2005). The small range of binding energy and a lack of critical energies observed in the experimental data make a detailed discussion of this discrepancy impossible.

Table 3 lists the Fermi wave vectors and Fermi velocities from experiment (Schäfer et al 2005) for each of the theoretical models considered the obtained mass renormalization values. Comparison of the Fermi velocities is done via the mass renormalization, which is calculated as the ratio of theoretical to experimental Fermi velocity. Comparison of the Fermi vectors indicates that the average difference between experiment and theory...
Table 3. Fermi wave vectors, Fermi velocities and mass renormalization values ($v_F,\text{theor}/v_F,\text{expt}$) determined by comparison of experimental values with ISM, ISM + SO, DTM and BTM.

| Direction (spin) | Fermi vector (Å$^{-1}$) | Fermi velocity (eV Å) | Mass renorm. | ISM (Schäfer et al 2005) | ISM + SO |
|------------------|-------------------------|-----------------------|-------------|--------------------------|----------|
|                  |                         |                       |             |                          |          |
| $\Gamma$–P       | VI                      | 0.32                  | 0.88        | 0.3                      | 1.73     | 1.97     | 0.25    | 1.55    | 1.76    |
|                  | I                       | 0.97                  | 1.4         | 0.94                     | 5        | 3.57     | 0.94    | 4.23    | 3.02    |
| $\Gamma$–N       | VI                      | 0.36                  | 0.8         | 0.32                     | 1.47     | 1.84     | 0.26    | 1.45    | 1.81    |
|                  | I                       | 1.22                  | 1.16        | 1.21                     | 2.37     | 2.04     | 1.16    | 2.38    | 2.05    |
| $\Gamma$–H       | VI                      | 0.46                  | 0.72        | 0.46                     | 1        | 1.39     | 0.35    | 0.54    | 0.75    |
|                  | VII–V                  | 1.18                  | 1.12        | 1.02                     | 1.18     | 1.05     | 1.08    | 1.39    | 1.24    |
|                  | I                       | 1.08                  | 1.12        | 1.05                     | 3.78     | 3.38     | 1.05    | 3.01    | 2.69    |
|                  | II                      | 1.7                   | 0.67        | 1.9                      | 0.72     | 1.07     | 1.84    | 0.71    | 1.06    |
| $H$–P            | V                       | 0.68                  | 1.79        | 0.66                     | 2.94     | 1.64     | 0.65    | 1.55    | 0.87    |

| Direction (spin) | Fermi vector (Å$^{-1}$) | Fermi velocity (eV Å) | Mass renorm. | DTM | BTM |
|------------------|-------------------------|-----------------------|-------------|-----|-----|
|                  |                         |                       |             |     |     |
| $\Gamma$–P       | VI                      | 0.25                  | 1.55        | 1.76| 0.27| 1.33| 1.51 |
|                  | I                       | 0.93                  | 4.17        | 2.98| 0.97| 3.14| 2.24 |
| $\Gamma$–N       | VI                      | 0.27                  | 1.42        | 1.78| 0.32| 0.79| 0.99 |
|                  | I                       | 1.13                  | 2.81        | 2.42| 1.16| 1.31| 1.13 |
| $\Gamma$–H       | VI                      | 0.36                  | 0.52        | 0.72| 0.39| 0.81| 1.13 |
|                  | VII–V                  | 1.08                  | 1.36        | 1.21| 1.03| 1.16| 1.04 |
|                  | I                       | 1.05                  | 3.02        | 2.7 | 0.95| 1.41| 1.26 |
|                  | II                      | 1.82                  | 0.71        | 1.06| 1.79| 0.81| 1.21 |
| $H$–P            | V                       | 0.65                  | 1.56        | 0.87| 0.69| 1.97| 1.10 |

$^1$The mass renormalization is taken as the ratio of theoretical to experimental Fermi velocity as is common.
decreases the more exhaustive the theoretical model becomes. Although the effects of spin–orbit coupling, transitions into final states and broadening on the Fermi vectors are rather small, these effects are more pronounced when considering the Fermi velocities, i.e. the mass renormalization values. Figure 4 indicates that mass renormalization decreases substantially when increasing the integrity of the theoretical treatment (moving from left to right in table 3 and figure 4) in 80% of the Fermi level crossings considered. When assessing the whole set of sampled \( k \)-points, it is seen that the mass-renormalization values are overestimated in Schäfer et al (2005) by 60% on average. Instead of an average mass renormalization of 2.3, we obtain a value of 1.4. Thus, inclusion of spin–orbit coupling, initial to final state transitions and broadening significantly increases the accuracy of the theoretical description of the experimental data.

6. Discussion

We have seen that the recently reported large bandwidth renormalizations are mostly an effect of the photoemission measurement and of the choice of a semi-relativistic initial state. This is in agreement with literature data, which lead to a bandwidth reduction of only \( \sim 10\% \) without a conceivable trend towards enhancement for shallow bands (Rader and Gudat 1999).
It is, however, problematic for the analysis that scattering between different LDA calculations increases near $E_F$ (Rader and Gudat 1999). Concerning the comparison of the Fermi velocity, which is also based on a larger number of experimental data points than the bandwidth (Schäfer et al 2005), we find with 1.4 on average a considerably smaller mass renormalization than Schäfer et al but within the limits of previous de Haas–van Alphen results (Lonzarich 1984). There are not many studies on the Fermi velocity of metals by photoemission. For Ni, a large mass renormalization of $v_F$ of 1.9–2.8 has recently been reported (Higashiguchi et al 2005). It was found to be in agreement with both de Haas–van Alphen data and the established bandwidth reduction in Ni of $\sim 30\%$. The mass normalizations that were before (Schäfer et al 2005) in part found in the upper range of the values inferred from de Haas–van Alphen data (Lonzarich 1984) are now well inside the rather broad range of de Haas–van Alphen values of 1.5–3 (Lonzarich 1984). The mass renormalization of $v_F$ in Ni is somewhat smaller for enhanced sp-character in de Haas–van Alphen (Zornberg 1970) and photoemission data (Higashiguchi et al 2005). In fact, mass renormalization of the sp-states in Ag is small as derived from de Haas–van Alphen data (1.05) and photoemission from quantum-well states (0.96) (Hong et al 2002).

Spin-dependent scattering has been suggested (Fedorov et al 2002, Hong and Mills 1999, Hong et al 2002) to lead to increased mass renormalization in either majority- or minority-spin bands. This is not observed for the current analysis, with the two highest values, 2.24 and 1.51, found on bands of opposite spin. A possible mechanism relating the proximity of opposite spin sheets of different sizes to high mass renormalization (Schäfer et al 2005) is not supported by the current analysis since our highest mass renormalization value, 2.24, remains on sheet I along $\Gamma P$ with no sheet of opposite spin nearby.

Spin-wave renormalization (electron–magnon interactions) and electron–phonon interactions of bulk (Cui et al 2006, 2007) and surface (Schäfer et al 2004) states in ferromagnetic iron have been observed and may be responsible for the additional renormalization; comparison of sheet I ($k_F = 0.95 \, \text{Å}^{-1}$) in figure 2 and the corresponding experimental band (see Schäfer et al (2005); figure 12) indicates that the deviation between experiment and theory continues down to at least 0.5 eV binding energy. This is significantly larger than expected from magnon (surface, 0.125–0.16 eV (Schäfer et al 2004), and bulk, 0.270 eV (Cui et al 2006, 2007)) and phonon (bulk, 0.04 eV (Cui et al 2006, 2007)) renormalization. Such interactions are observed as kinks or deviations from the predicted band structure. In the vicinity of the kink, these deviations amount, in the vicinity of the kink, to $<0.02 \, \text{Å}^{-1}$ in the case of coupling to magnons (Cui et al 2006, 2007, Schäfer et al 2004) and $<0.004 \, \text{Å}^{-1}$ for coupling to phonons (Cui et al 2006, 2007), while the current variation is an order of magnitude larger ($\sim 0.15 \, \text{Å}^{-1}$); a similar deviation is observed for the other bands where mass renormalization is observed. It is therefore believed that the mass renormalization observed in the current data is the result of many electron interactions as the deviation between experiment and theory is significantly larger than expected for electron–magnon and electron–phonon interactions.

7. Conclusion

Recently, large correlation-induced renormalizations of the occupied bandwidth and the Fermi velocity of ferromagnetic iron based on DFT of initial states were reported. We have applied a similar DFT for initial states and three models of angle-resolved photoemission. The most comprehensive model is a broadened transition model that incorporates the calculation of spin–orbit coupling, initial to final state transitions and broadening effects into the DFT of
initial states in the three-step model of photoemission. The Fermi vector, mass renormalization and bandwidth renormalization values obtained from this model give greater agreement with experimental data than density functional calculations (Schäfer et al 2005) of the initial states alone. The bandwidth renormalization within 1 eV of the Fermi energy is not found to be $\sim 30\%$ as in Ni but of the order of 10%. This is in contradiction to recent LDA + DMFT calculations (Grechnev et al 2007), which indicated that the real component of the self-energy would provide a bandwidth renormalization of $\sim 50\%$; a detailed investigation of this discrepancy is left to future work including a larger number of critical point energies over a wider binding energy range. Neither a preferred scattering for minority spin as found for Gd (Fedorov et al 2002) nor for majority spin as predicted by spin-flip scattering under the emission of a spin wave (Hong and Mills 1999) is found here, in agreement with the previous analysis (Schäfer et al 2005). No obvious spin dependence of the mass renormalization is found here, and the vicinity of Fermi surface sheets of opposite sign and much smaller occupied fraction does not enhance the mass renormalization. The increased mass renormalization is attributed to many-electron interactions due to the large energy (0.5 eV) and momentum ($\sim 0.15 \text{ Å}^{-1}$) deviations between experiment and theory. It is to the merit of Schäfer et al that they have pointed out the persistence of a substantial mass renormalization in ferromagnetic transition metals, an effect that has largely been ignored since the de Haas–van Alphen results (Lonzarich 1984). It is shown in this paper that while photoemission enables detailed $\mathbf{k}$-dependent investigations, the size of the renormalization is reduced by a better description of the photoemission process. A description with improved accuracy is achieved within the three-step model of photoemission without significant computational complexity. In this context, it would be desirable to verify the present results on final-state effects on the Fermi velocity also within a one-step model (Braun 1996, Redinger et al 1988) and using advanced theoretical methods treating electron correlation such as dynamical mean field theory (Lichtenstein et al 2001).

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