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Angle-resolved photoemission from Fe₃Pt: theory and experiment

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Abstract. Angle-resolved photoelectron spectra have been measured at room temperature (i.e. well below the Curie temperature) from the (001) surface of an Fe₃Pt single crystal for a variety of emission angles using unpolarised 21.2 eV radiation. Corresponding spectra were calculated by means of a one-step theory of photoemission together with the bulk band structure and the one-dimensional DOS. The fairly good overall agreement reached between theory and experiment firstly provides an explanation of the observed features in terms of direct bulk inter-band transitions from majority and minority spin states and secondly favours, amongst local density exchange–correlation approximations, an X_α version with $\alpha = 0.7$ around 2 eV below E_F and a somewhat smaller α closer to E_F .

1. Introduction

In line with the persisting interest in magnetovolume effects (cf Capellmann 1987 and Wassermann 1989, and references therein), FCC ordered Fe₃Pt, a typical Invar system, has recently been studied in a pioneering spin-resolved photoemission experiment (Carbone *et al* 1987, Kisker *et al* 1987). Spectral changes observed in going towards and beyond the Curie temperature could be explained by a correlated-local-moment theory of finite-temperature photoemission in terms of changes in the total densities of states (Gollisch and Feder 1989). As the data involved a large amount of k_{\parallel} -integration, conclusions on details of the electronic structure were, however, not possible.

Since an understanding of the electronic structure at high temperatures first requires knowledge of it at low temperature, we have performed for Fe₃Pt angle-resolved photoemission measurements at room temperature (i.e. well below the Curie temperature of 550 K) for a variety of emission angles and have subsequently analysed the data theoretically by calculations of the spin-dependent band structures, densities of states and one-step-model photoemission spectra.

The experimental and theoretical procedures are outlined in sections 2 and 3, respectively. Results are presented and discussed in section 4.

2. Experiment

The instrumental equipment used in these experiments has been described previously (Heimann *et al* 1979). It consists of a measurement and a preparation chamber, which are connected via a gate valve. The electron energy analyser has an energy resolution of better than 60 meV for the photoemission experiments (full width at half maximum). The angle resolution is $\pm 2^\circ$ (full angle of the acceptance cone). The photoelectrons are excited by unpolarised rare gas resonance radiation. The angular resolved spectra are obtained by rotating the sample in front of the entrance slit of the energy analyser; the angle between incident light and electron emission being always constant (approximately 80°). Cleanliness of the sample and concentration of the constituents are controlled by Auger electron spectroscopy (AES), and surface order by low-energy electron diffraction (LEED).

The sample was prepared by melting the components under argon atmosphere, subsequent homogenisation by annealing and crystallisation by the Bridgman technique. The bulk concentration of the constituents in Fe_3Pt was confirmed by x-ray fluorescence analysis, the crystalline order by x-ray diffraction. The (100) sample was obtained by orienting the Fe_3Pt single crystal within an accuracy of $\pm 1^\circ$ using the Laue pattern, cutting by means of a diamond saw, mechanical grinding and polishing and subsequent electrolytic polishing. *In situ* preparation consisted of numerous cycles of Ar (and finally Ne) ion bombardment (at energies of approximately 600 eV) and subsequent annealing to a temperature of less than 600°C until surface contamination could no longer be detected in the AES spectra. No surface enrichment of one alloy component by preferential sputtering or segregation was found. LEED in the final state showed a clear (100) pattern without indication of a surface reconstruction. Experiments have been performed both by using initially ordered and disordered Fe_3Pt samples (as analysed by x-ray diffraction). Both kinds of sample gave rise to the same LEED pattern, which means that the necessary *in situ* treatment of the samples always led to ordering in the surface region.

3. Theory

The general foundation is an effective quasi-particle Hamiltonian for the semi-infinite magnetic crystal (cf Feder 1985 and references therein), which contains different complex self-energy parts for the hole (lower) states and the photoelectron (upper) states. The large atomic number of Pt ($Z = 78$) might recommend a fully relativistic treatment. However, the Pt-derived d bands are well separated in energy from the Fe-derived d bands (Hasegawa 1985, Podgorny 1989). The former are below -3.5 eV, and photoemission features from them are therefore strongly broadened by hole-life-time effects and masked by the inelastic background. The Fe-derived d bands are between -3.5 eV and the Fermi energy and are therefore responsible for practically all the features in the present photoemission spectra. It is, therefore, warranted to avoid the substantial additional computational labour brought about by spin-orbit coupling and to adopt a one-component scalar-relativistic form. For the magnetic part of the Hamiltonian, we neglect—working well below the Curie temperature—the fluctuations of local moments and adopt a Stoner-like model, in which the same effective magnetic field resides on all Fe sites. This renders all unit cells equivalent and completely separates the problem into a majority- and a minority-spin part with corresponding effective potentials (cf e.g. Feder 1985).

Consequently, photoemission can be treated—in the spirit of a one-step model in the electric dipole approximation—by a non-relativistic multiple scattering formalism developed by Pendry (1976). Since Fe_3Pt has four atoms per unit cell, the usual one-atom version (Hopkinson *et al* 1980) has to be replaced by a several-atom one (Pendry and Blake 1988). The near-grazing incidence of the photons in the present experiment requires consideration of the effect of the surface on the photon field. We do this by employing inside the solid a classical electromagnetic field as determined from the incident field by Snell's law and Fresnel's equations, which for mainly direct inter-band transitions was found to be adequate by Goldmann *et al* (1983). As we are not aware of any determination of the refractive index of Fe_3Pt , we resort to optical data for Fe and Pt (Weaver *et al* 1981), which gives, at the present photon energy of 21.2 eV, the complex value $0.79 + 0.56i$. In view of identifying the physical origin of individual photoemission features, we also calculate, for the real parts of the potentials used in the photoemission calculations, the lower and upper bulk band structures along Γ -X and the corresponding one-dimensional densities of states.

It remains to specify the spin-dependent effective quasiparticle potential. In the absence of first-principles results, we follow a procedure established by previous photoemission work (Feder *et al* 1984, Feder 1985), which starts from majority- and minority-spin charge densities of the ground state. We take these from a recent self-consistent scalar-relativistic LMTO calculation (Podgorny 1989), in which exchange and correlation were treated in the local spin-density approximation of Vosko *et al* (1980). However, caution is indicated in two respects. Firstly, the lattice constant at minimum total energy found by Podgorny (1989) is 0.09 \AA smaller than the experimental value of 3.75 \AA . Since this turns out to imply a lowering of the relevant upper bands by about 1 eV, we also use charge densities obtained by renormalising the original ones to the larger unit cell. Secondly, the magnetic moment per unit cell obtained from the majority and minority charge densities in Podgorny (1989) ($7.8 \mu_B$) is about 15% smaller than an experimentally determined one ($9.2 \mu_B$) (Ishikawa *et al* 1980). As an alternative we therefore employ charge densities obtained from Podgorny (1989) by increasing (reducing)—with the same weight at each radial point r —the majority (minority) densities so as to produce the larger magnetic moment. We note that this is in line with the well known trend (found e.g. for FCC Fe by Moruzzi *et al* (1986, 1989), Krasko (1987) and references therein) that an increase of the lattice constant entails an increase of the magnetic moment.

In constructing from the above charge densities the (spherically symmetric) potential parts inside the muffin-tin sphere we use as local-density exchange-correlation approximations the one due to Vosko *et al* (1980) and the X_α prescription with various values of α . The idea behind this 'openness' is that the prescription favoured by comparison between theoretical and experimental photoemission spectra should to some extent mimic non-local exchange-correlation and real self-energy corrections. The complex uniform inner potential is chosen as follows. For its real part for the initial states we take, according to the usual form $\phi + E_F$, 13.6 eV. Small changes for the various charge density and exchange-correlation options discussed above have practically no effect for normal emission and little for off-normal. For the final states we firstly use the same value and secondly a value reduced by about 1 eV 'self-energy corrections' as suggested by experience with LEED (cf e.g. Feder 1985). The imaginary part of the initial state self-energy is chosen in the form $pE^2/(E^2 + E_0^2)$ with parameter values $p = 2 \text{ eV}$ and $E_0 = 5 \text{ eV}$ (cf Gollisch and Feder 1989). For the final states we use 1 eV on the grounds of experience with LEED (Feder 1985).

4. Results and discussion

In order to explore the origin of photoemission features from Fe₃Pt, we first show in figure 1 calculated normal-emission spectra together with the Δ_1 and Δ_5 bulk bands along Γ -X and the associated one-dimensional densities of states, as obtained from the charge densities of Podgorny (1989) modified to the experimental lattice constant and magnetic moment (cf above) and using the Vosko *et al* (1980) exchange-correlation approximation.

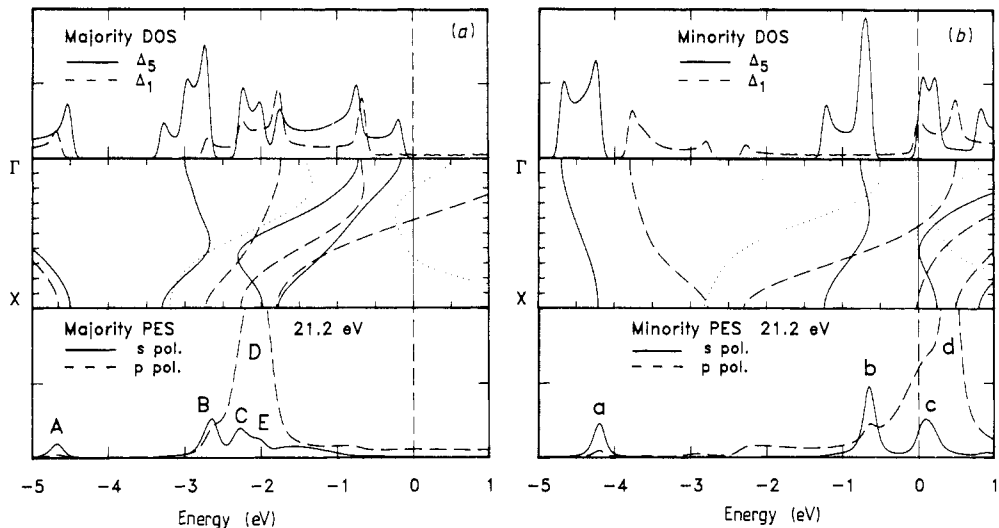


Figure 1. (a) Fe₃Pt(001) majority spin (panels from top to bottom): one-dimensional density of initial states of symmetry Δ_5 (full curve) and Δ_1 (broken curve); initial-state Δ_5 (full curve) and Δ_1 (broken curve) bands, and final-state Δ_1 bands (dotted curve) shifted downward by the photon energy 21.2 eV; normal photoemission spectra from Δ_5 (full curve) and Δ_1 (broken curve) semi-infinite-crystal states. (b) As (a), but for minority spin.

For majority spin (figure 1(a)), the predominantly Fe-like d bands are seen to lie between -3.3 eV and E_F . The dominant photoemission peak, labelled D, is produced by p-polarised light, i.e. from a Δ_1 band by A_z (the surface-normal component of the vector potential \mathbf{A}). Since its energy position is at the crossing point of the occupied Δ_1 band with a final-state Δ_1 band (dotted curve) displaced downward by the photon energy, it can be interpreted as a direct bulk inter-band transition. The photoemission structures produced by s-polarised light, i.e. from Δ_5 initial states, are seen to be much weaker and correlated with maxima in the DOS. With the exception of B, which is due to a direct transition, they are ascribed to excitation into final states (time-reversed LEED states) that are evanescent inside the solid. For minority spin (figure 1(b)), the Fe-like d bands are shifted upward by an exchange splitting of about 2.3 eV and the Fermi energy goes through them. The exchange splitting of the final-state bands is only about 0.5 eV. For p-polarised light, the dominant feature, labelled d, is at about 0.4 eV above E_F . A contribution to the photocurrent comes therefore only from the shoulder around E_F , which can be interpreted as a DOS transition into evanescent states. For s-polarised light, the main feature b at -0.7 eV is interpreted as due to

direct transition. The slight displacement in energy from the crossing point of the lower Δ_5 band and the (downward shifted) upper Δ_1 band is due to the latter having been calculated for the real part of the potential, whilst strictly speaking one should employ the complex band as obtained from the complex upper-state potential. The separation of about 2.1 eV between the majority peak B and the minority peak b can be regarded as an observable exchange splitting, but one has to bear in mind that the two initial-state Δ_5 bands are not rigidly exchange split and that the underlying direct transitions occur at different points in the Brillouin zone.

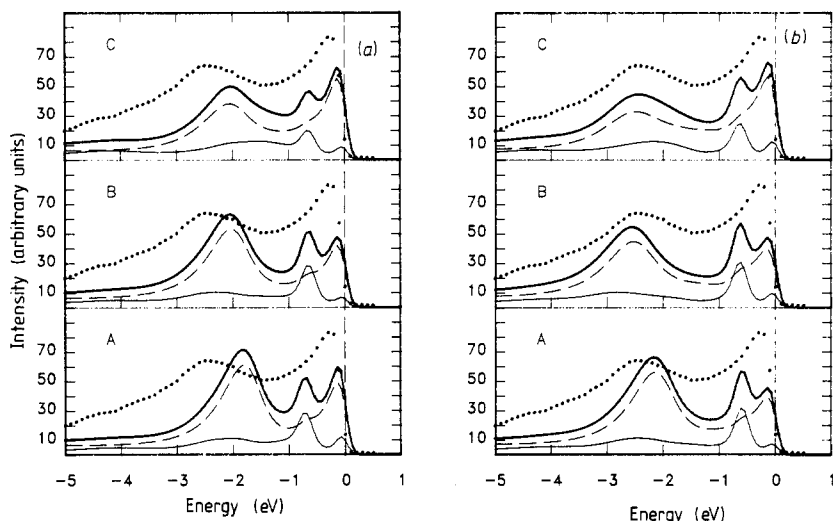


Figure 2. (a) Normal photoemission spectra from Fe_3Pt (001) by 21.2 eV photons incident at 80° with respect to surface normal: experiment with unpolarised light (dotted curve) and theory with Vosko *et al* (1980) exchange–correlation approximation for s-polarised light (thin full curve), p-polarised light (thin broken curve) and sum (thick full curve), for magnetic moment per unit cell $7.8\mu_B$ (from Podgorny 1989) (bottom panel), $9.2\mu_B$ (central panel), and final-state real self-energy correction -1.2 eV (top panel). (b) As (a), but theoretical spectra for X_α exchange with $\alpha = 0.7$.

Figure 2 firstly shows the experimental spin-summed normal-emission spectrum produced by unpolarised 21.2 eV radiation incident at 80° to the surface normal. It is seen to consist mainly of a leading maximum near E_F and a broader peak around about -2.5 eV. From figure 1 it is clear that the former has minority and the latter majority spin. Comparison is made with spin-summed spectra calculated for s- and p-polarised light for the various potentials described in section 3. The spectra in figure 2(a) were obtained with the Vosko *et al* (1980) exchange–correlation approximation with magnetic moment $7.8\mu_B$ (bottom panel) and $9.2\mu_B$ (upper two panels). For the latter, the peak near -2 eV, which is mainly the direct-transition majority feature D (in the notation of figure 1(a)), is shifted to lower energy by about 0.2 eV. For a final-state real self-energy correction of -1.2 eV, which implies a 1.2 eV upward shift of the final-state bands shown in figure 1, the top panel of figure 2(a) shows that the -2 eV peak is marginally shifted to lower energy (in accordance with the displacement of the crossing point of the bands in figure 1(a)) and that its height relative to the peak near E_F is significantly reduced. The theoretical spectra in figure 2(a) reproduce

the two experimental peaks with a discrepancy in position of about 0.5 and 0.2 eV. In addition, they exhibit a peak near -0.7 eV, which is identified from figure 1(b) as the minority Δ_5 derived feature b.

Analogous theoretical spectra calculated with X_α exchange with $\alpha=0.7$ are shown in figure 2(b). For the larger magnetic moment and the final-state real self-energy correction -1.2 eV (top panel), the majority peak is now at the same energy as in the experimental spectrum, but the discrepancy around -0.4 eV remains. The band structure in figure 1(b) indicates that it can be cured by shifting the relevant minority Δ_5 band by about 0.3 eV towards E_F . Such a shift could be produced *ad hoc* by employing an energy-dependent α that mimics the real part of the initial-state self-energy (cf Feder *et al* 1984).

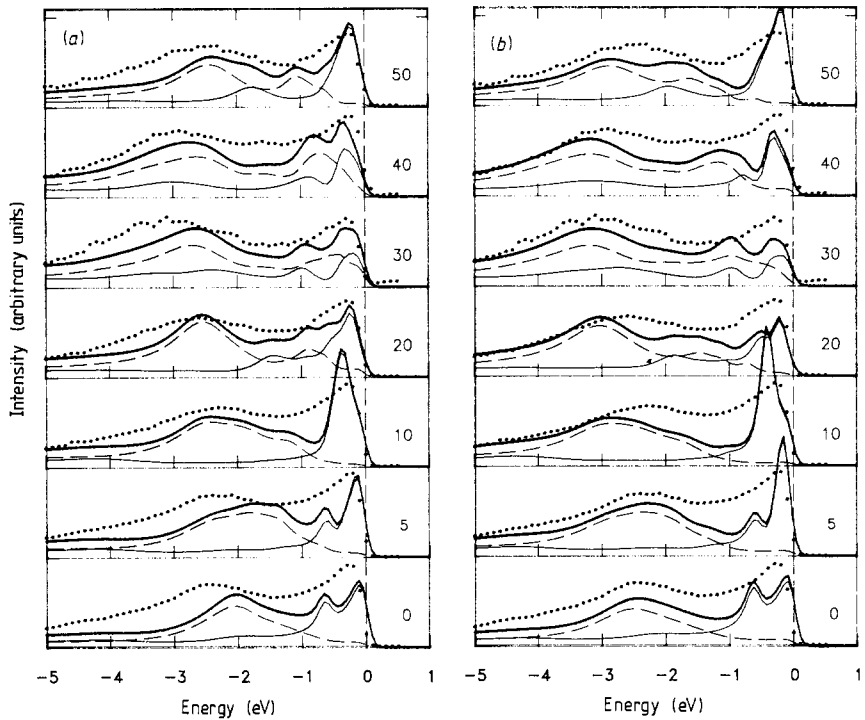


Figure 3. (a) Photoemission spectra from $\text{Fe}_3\text{Pt}(001)$ at angles θ_e as indicated in the panels by unpolarised 21.2 eV photons incident at $\theta_p = 80^\circ - \theta_e$: experiment (dotted curve) and theory with Vosko *et al* (1980) exchange for majority spin (thin broken curve), minority spin (thin full curve) and spin-sum (thick full curve). (b) As (a), but theory for X_α exchange with $\alpha = 0.7$.

A sequence of photoelectron spectra for emission angles between 0° and 50° has been measured and is compared to theoretical results obtained with the Vosko *et al* (1980) exchange–correlation approximation (figure 3(a)) and the X_α approximation with $\alpha = 0.7$ (figure 3(b)). With increasing emission angle, the experimental majority-spin feature, which is at -2.5 eV for $\theta = 0^\circ$, is seen to move slightly upward until 10° , downward until 30° and then upward again. This behaviour is reproduced by both sets of calculated spectra, with the X_α majority features rigidly displaced downward by about 0.5 eV with regard to their Vosko-type counterparts. With regard to the

energy position of the majority feature, comparison with experiment favours X_α for near-normal emission and 30° , Vosko for 10° and 20° , and something in between for 40° and 50° .

The minority-spin feature around 0.3 eV in the experimental spectra shows practically no dispersion and acquires a small shoulder around 0.6 eV for the larger angles. Both sets of theoretical spectra exhibit two peaks above -1 eV, one near 0.1–0.2 eV developing from the normal-emission features c and d (cf figure 1(b)) and one near 0.7 eV developing from b. We note the strong changes of these features with angle between 0 and 20° . The much weaker changes in the experimental spectra can partly be understood from the fact that they involve some angular integration, but further indicate that the band developing from the flat Δ_5 minority band (cf figure 1(b)) should be closer to E_F . This would also shift the theoretical features near -1 eV at $\theta = 30^\circ$ and 40° toward closer agreement with experiment.

The theoretical spectra, which we have shown, were obtained for an ideal Fe_3Pt (001) surface with an unrelaxed planar FePt layer on top followed by an alternating sequence of pure Fe and FePt layers. Calculations for the pure Fe layer on top yielded photoemission spectra practically identical to the former ones. This supports our above understanding of the main spectral features in terms of bulk inter-band transitions. If an appreciable contribution to the spectra originated from the outermost layer of atoms, spectra calculated for the two termination cases should differ appreciably from each other. The absence of such differences suggests also an insensitivity of the spectra to a possible buckling of the topmost FePt layer.

5. Conclusion

Angle-resolved photoemission spectra measured from Fe_3Pt can be interpreted in terms of direct bulk inter-band transitions originating from majority- and minority-spin states. Fairly good overall agreement with the experimental spectra has been obtained by one-step-model photoemission calculations starting from an *ad hoc* modified self-consistent spin-dependent charge density and assuming for the exchange correlation part of the self-energy an X_α approximation with $\alpha = 0.7$ rather than the RPA local-density approximation due to Vosko *et al* (1980). Assuming that an energy dependence of the self-energy improves the agreement close to E_F , but implies altogether too many fit parameters to be physically meaningful at this stage. Since the calculated spectra are quite sensitive to details of the underlying charge density and of the exchange–correlation approximations, presently remaining discrepancies between theory and experiment can, however, be viewed as promising further physical insight if further work is done simultaneously in the following three directions. Firstly, self-consistent ground state calculations should be performed with improved exchange–correlation approximations, e.g. including gradient corrections recently found successful by Bagno *et al* (1989) for third-row elements. Secondly, the experimental basis should be enlarged by resolution with respect to electron spin and light polarisation. Thirdly, such data should be analysed by photoemission calculations employing refined energy-dependent approximations to the exchange–correlation parts of the self-energies of the photoelectron and of the quasi-hole.

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References

- Bagno P, Jepsen O and Gunnarsson O 1989 *Phys. Rev. B* **40** 1997
Capellmann H 1987 (ed) *Metallic Magnetism* (Berlin: Springer) ch 6
Carbone C, Kisker E, Walker K H and Wassermann E F 1987 *Phys. Rev. B* **58** 1784
Feder R (ed) 1985 *Polarized Electrons in Surface Physics* (Singapore: World Scientific) ch 4
Feder R, Rodriguez A, Baier U and Kisker E 1984 *Solid State Commun.* **52** 57
Goldmann A, Rodriguez A and Feder R 1983 *Solid State Commun.* **45** 449
Gollisch H and Feder R 1989 *Solid State Commun.* **69** 579
Hasegawa A 1985 *J. Phys. Soc. Japan* **54** 1477
Heimann P, Hermanson J, Miosga H and Neddermeyer H 1979 *Phys. Rev. B* **20** 3059
Hopkinson J F L, Pendry J B and Titterton D J 1980 *Comput. Phys. Commun.* **19** 69
Ishikawa Y, Tajima K, Noda Y and Wakabayashi N 1980 *J. Phys. Soc. Japan* **48** 1097
Kisker E, Wassermann E F and Carbone C 1987 *Phys. Rev. Lett.* **58** 1784
Krasko G L 1987 *Phys. Rev. B* **36** 8565
Moruzzi V L, Marcus P M and Kübler J 1989 *Phys. Rev. B* **39** 6957
Moruzzi V L, Marcus P M, Schwarz K and Mohn P 1986 *Phys. Rev. B* **34** 1784
Pendry J B 1976 *Surf. Sci.* **57** 579
Pendry J B and Blake R 1988 private communication
Podgorny M 1989 *Physica B* **161** 110; 1989 private communication
Vosko S H, Wilk L and Nusair M 1980 *Can. J. Phys.* **58** 1200
Wassermann E F 1989 *Phys. Scr.* **25** 209
Weaver J H, Krafka C, Lynch D W and Koch E E 1981 (ed) *Optical Properties of Metals II* (internal report DESY F41)