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Momentum profiles of aluminum

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Abstract

Electron momentum spectroscopy is a tool ideally suited for the study of the energy-resolved momentum densities. Here we present new data from a high-energy EMS experiment using 50 keV incoming and 25 keV outgoing electrons. Momentum profiles have been measured from a thin aluminum film for the 2*p* core level and the valence band near E_f. The resolution of these momentum profiles, as well as the effects of multiple scattering are discussed. It is found that the high-energy EMS experiment measures momentum densities with superior resolution and very little background.

Keywords: Electron momentum spectroscopy; (*e*, 2*e*); Spectral function; Aluminum; Momentum density

1. Introduction

For the study of the electronic structure of crystalline solids photo-emission is the most widely used tool. An incoming photon is absorbed by the system, and an electron is emitted. The momentum of the emitted electron is much larger than that of the incoming photon, and hence the difference in momentum of the initial and final state is compensated by phonons [1].

It is however possible for an incoming particle (photon or electron) to have a binary collision with a target electron only. The scattered and ejected particles are emitted with energies and momenta, as dictated by the laws of conservation of energy and momenta. The remainder of the target is essentially a spectator observing the annihilation of one of its electrons. In this case the target need not be a single crystal but may even be amorphous. The most familiar technique using this type of collisions is Compton scattering in which case only the energy of the scattered photon is measured [2]. As a consequence of this only a projection of the momentum vector is resolved in this technique. Using incident electrons it is possible (although not easy) to measure in coincidence the scattered electron as well as the ejected one. The technique is then referred to as (*e*, 2*e*) spectroscopy or electron momentum spectroscopy (EMS) [3–5]. Analysing both particles for energy and momentum allows for the determination of the binding energy and momentum of the ejected electron prior to the collision event.

Due to the simplicity of the binary collision process one obtains very direct information about the spectral momentum density. The scattering cross section is just the Mott cross section for (free)

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electron–electron collisions [3]. Using suitable kinematics the observed intensity in an EMS experiment is then directly proportional to the target electron spectral momentum density.

A horizontal cross section of the current spectrometer is shown in Fig. 1 [6,7]. The electron gun produces 25 keV electrons. The thick lines indicate the positive high voltage area (25 keV), containing the sample consisting of a suitable mounted thin self-supporting film, the dotted lines correspond to the electron optics and analyzers. Thus, 50 keV electrons impinge on the sample. The emerging electrons with energies near 25 keV are then detected in coincidence in the two electrostatic analyzers (at polar angle $\theta = 44.3°$) that are near ground potential. From each electron the azimuthal angle $\phi_{1,2}$ and energy $E_{1,2}$ are determined from which the separation energy and the target recoil momentum are determined. The two electrostatic analysers have slit-like openings that extend along the $\theta = 44.3°$ cone in the vertical direction (perpendicular to that of Fig. 1). The angle θ is chosen such that, if the incoming and outgoing trajectories are all in the same plane ($\phi = \phi_1 - \phi_2 - \pi = 0$), we scatter from a stationary electron. If $\phi \neq 0$, i.e. the ejected electron is outside the plane defined by the momentum of the incoming and scattered electron, the ejected electron before the collision had a momentum proportional to ϕ, directed along the vertical direction. Thus, we can measure simultaneously a range of target momenta.

For comparison we will compare our data with results from the Flinders university spectrometer [8]. This spectrometer is used to produce the highest quality EMS data of solids. It has an asymmetric geometry with incoming electrons at an energy of 20 keV, and measures outgoing electrons at energies of 18.8 and 1.2 keV. The relatively low energy of the 1.2 keV electron was the main cause of rather severe multiple scattering in this design.

An example of the raw measurement data as obtained with the new spectrometer is shown in Fig. 2 for a thin aluminum film evaporated on amorphous carbon. As the aluminum film is thicker ($\simeq 50$ Å) than the 30 Å thick carbon film, the main features are from aluminum. We see a parabola at small binding energy (energies are relative to the vacuum level, the Fermi level is at about 5 eV binding energy.) as expected for a free-electron solid, such as aluminum. At high binding energy ($\simeq 78$ eV) there is a weak feature that has a binding energy in-

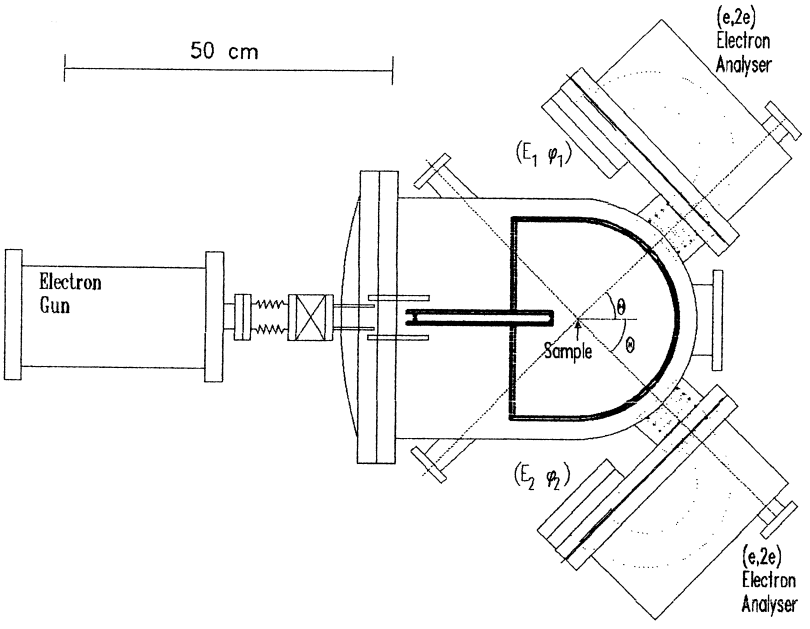

Fig. 1. The outline of the spectrometer. The thick lines correspond to the positive high voltage area, the dotted line to the electron optics.

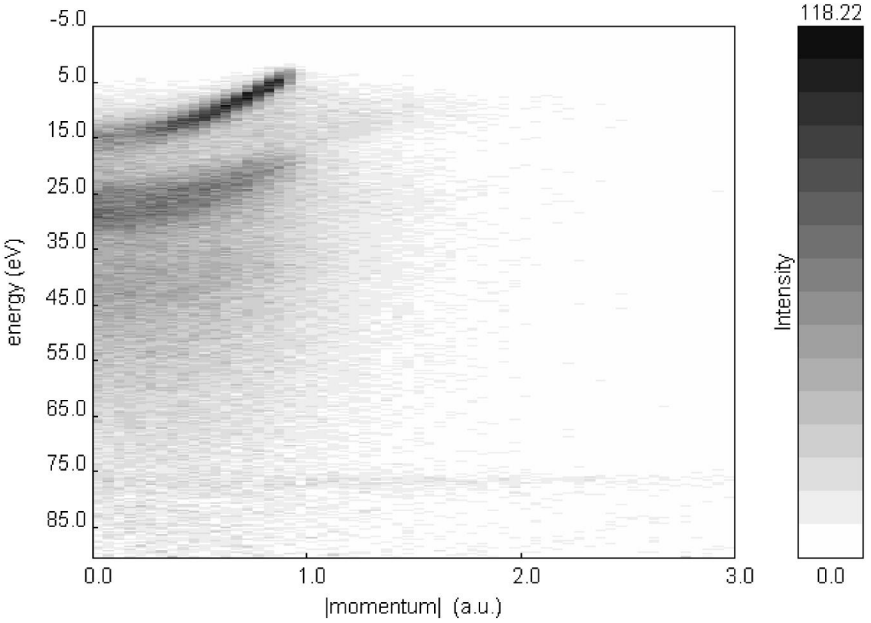

Fig. 2. The observed intensity plotted as a function of energy and momentum. The target was an approximately 50 Å thick Al evaporated on 30 Å carbon film. Both the Al valence band, resembling a free electron parabola and the Al 2*p* core level are visible.

dependent of momentum. This is the Al 2*p* core level. Thus, here we see two extreme cases in the same measurement, a contribution from a core level, with atomic characteristics showing no dispersion, and a contribution of nearly free-electrons. In this paper we want to compare the measured momentum densities with the calculated ones. From the observed intensity distribution in Fig. 2 it is clear that the (*e*, 2*e*) process is completely different from photoemission. Here the core level is the weak feature, overshadowed by the valence band whereas in photoemission it is vice-versa. The broad dispersive feature about 15 eV below the valence free-electron parabola is due to contributions from intrinsic and extrinsic plasmons [9] and the underlying carbon σ band. It will not be discussed further here.

Our final goal is to measure accurately the spectral function of aluminum, with a low level of multiple scattering [9]. In order to accomplish this, it will be necessary to remove the carbon by sputtering, and it is desirable as well to increase the energy resolution (currently slightly better than 2 eV). Here we want to discuss the performance of the new spectrometer as far as momentum resolution is concerned. For this we study two features that are hardly affected by the carbon backing, i.e. the Al 2*p* core level and the momentum density at the Fermi level. The carbon film has no sharp features at the Al 2*p* binding energy and at the Fermi level the density of states of this semi-metal is very small.

First let us concentrate on the Al 2*p* level. A 2*p* orbital is anti-symmetric with respect to the nucleus in real space, and therefore has a node at zero in momentum space. The quality of a momentum density measurement can be judged from the depth of this narrow minimum. The observed Al 2*p* feature is on a small background due to (*e*, 2*e*) events in the valence band region that have lost additional energy due to inelastic multiple scattering. The core level intensity is determined by fitting the peak on a sloping background. The peak position and width were determined from integrating the intensity over all momenta, and kept fixed while fitting the intensity integrated over momentum intervals of 0.2 a.u. The results are shown in Fig. 3 together with the modulus square of the (atomic) Al 2*p* wave function in momentum space. The general agreement is good, however the measured intensity at small momenta

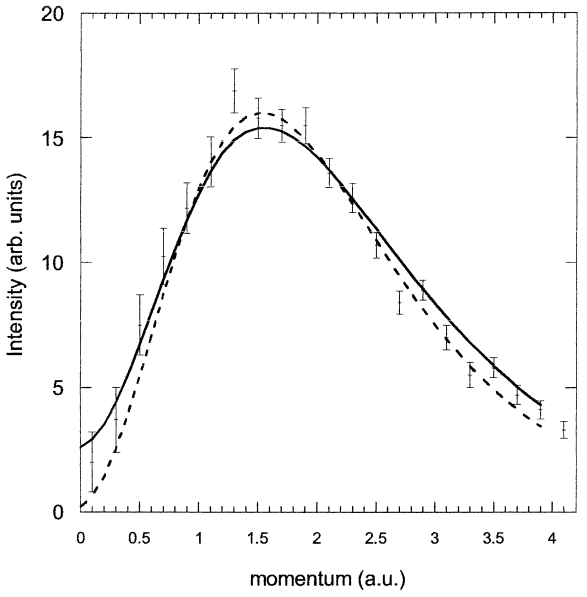

Fig. 3. The measured Al 2*p* momentum distribution. The dashed line is the modulus-square of the atomic Al 2*p* wave function. The full line is obtained using this wave function, but including the effects of elastic multiple scattering by the sample of the incident and emerging electrons by Monte Carlo simulations.

(< 0.5 a.u.) exceeds that given by the atomic wave function. There are three possible causes contributing to the measured non-zero intensity at zero momentum.

In the first place, the experiment has a finite momentum resolution. These experiments were done using the wide slits in front of each analyser (0.5 mm wide, 140 mm from the sample. The spectrometer can also operate with 0.2 mm slits, but this reduces the coincidence count rate significantly as it is proportional to the product of the two slit widths). In combination with the 0.1 mm diameter incident electron beam spot this results in a maximum variation of polar angle of ± 2.3 mrad. The 25 keV electrons have a momentum of 43.4 a.u., so ± 2.3 mrad corresponds to $\Delta p = \pm 0.1$ a.u. As one analyser is situated at $\simeq 90^\circ$ relative to the other, the Δp contribution of one analyser is (almost) perpendicular to that of the other. The combined momentum uncertainty thus has a maximum magnitude of ± 0.14 a.u. but the average error will be considerably smaller.

The third component of the momentum resolution vector is perpendicular to the plane of Fig. 1 and is determined by the azimuthal angular resolution of the analysers. The azimuthal angular resolution measured when small (0.1 mm diameter) pinholes are used to define the incoming beam is 1 mrad, hence the vertical momentum is measured with a maximum uncertainty of 0.04 a.u. As the perpendicular momentum is determined as the difference of two of these measurements its uncertainty will be about 0.06 a.u.

The collimation of the incoming beam is such that the spread of the momentum of the incoming electrons is even less. The excess intensity near zero momentum is thus unlikely to be entirely due to finite momentum resolution as the combined magnitude do not add up to the 0.3 a.u. required to explain the data.

The second possibility is an alignment error. The scattering angles θ were chosen in such a way that if the incoming and outgoing electrons are in the same plane we would have scattered from a stationary electron. A slight alignment error would result in scattering from an electron with a small momentum and hence larger intensity for the 2*p* level. Electrostatic deflectors along the emerging electron beams were used to mimic small rotations of both analysers. Here we used highly oriented pyrolytic graphite (HOPG) as a target as it increases the sensitivity of the test [10]. A small effect was found corresponding to a deviation of slightly less than 0.1 a.u. of the expected and measured zero momentum scattering conditions. The present measurements were done using the small correcting voltages on the deflectors determined in the graphite measurement. We expect the systematic error due to misalignment to be at most of the same magnitude as the angular resolution effects.

The third possibility is multiple scattering. If the incoming or an outgoing electron scatters from a nucleus it will transfer momentum rather than energy, and the inferred momentum of the ejected electron before the collision will be wrong, whereas the observed binding energy is not affected. These effects can be simulated using Monte Carlo simulations [11]. The full line in Fig. 3 is obtained from such a simulation for a 30 Å thick Al layer on top of a 30 Å thick C layer. This simulation accounts for all of the intensity at zero momentum, and this is thus

the most likely cause. Indeed the simulation appears to slightly overestimate the role of multiple scattering.

In comparison to similar experiments reported from the Flinders University EMS spectrometer [9] the present Al 2*p* profiles have a much deeper minimum at zero momentum. At Flinders the observed peak-to-valley ratio of the Al 2*p* core level was around 2, whereas in the present case this ratio is around 7.5. This also supports multiple scattering as being responsible for the filling in of the minimum, as the kinetic energy of the electrons is significantly larger in the present spectrometer (and hence the elastic cross sections are much smaller). On geometrical grounds alone we would not expect a large difference in momentum resolution between both spectrometers.

The full momentum resolving power of the spectrometer becomes obvious if we measure the momentum density near the Fermi level. For a free electron gas the momentum distribution at a certain energy would be a delta-function at k_f. The weak lattice potential will cause only small deviations away from this value level for some crystal orientations in our polycrystalline film. Moreover our finite energy resolution means that we sample the states near the Fermi level (spread over a small range of k values) as well (1 eV away from the Fermi level the k value is reduced by 0.05 a.u.). It is thus somewhat of a surprise that the measured intensity appears to have a width only slightly more than 0.1 a.u. These results are shown in Fig. 4 and compared with the earlier Flinders data [12]. Note that the uncertainty in the determination of the magnitude of k_f is now completely determined by the accuracy of the azimuthal angle measurement. The small momentum components due to the finite slit width (< 0.1 a.u) contribute negligibly to the magnitude of the momentum $|k|$ as they have to be added in squares to the large vertical component ($\simeq 0.9$ a.u.). Indeed measurements using the narrow slits results in similar momentum distributions.

The theory used to describe the Flinders measurement is broader than the present measurement and had to be re-evaluated on a finer momentum grid. The intrinsic width of the new calculation is about 0.05 a.u. The measured peak position appears agrees within 0.05 a.u. with the calculated ones and near perfect agreement is obtained if the effect of finite energy resolution is taken into account. Based on the electron density in Al the free electron model predicts a value of 0.93 a.u. for k_f, a value that is

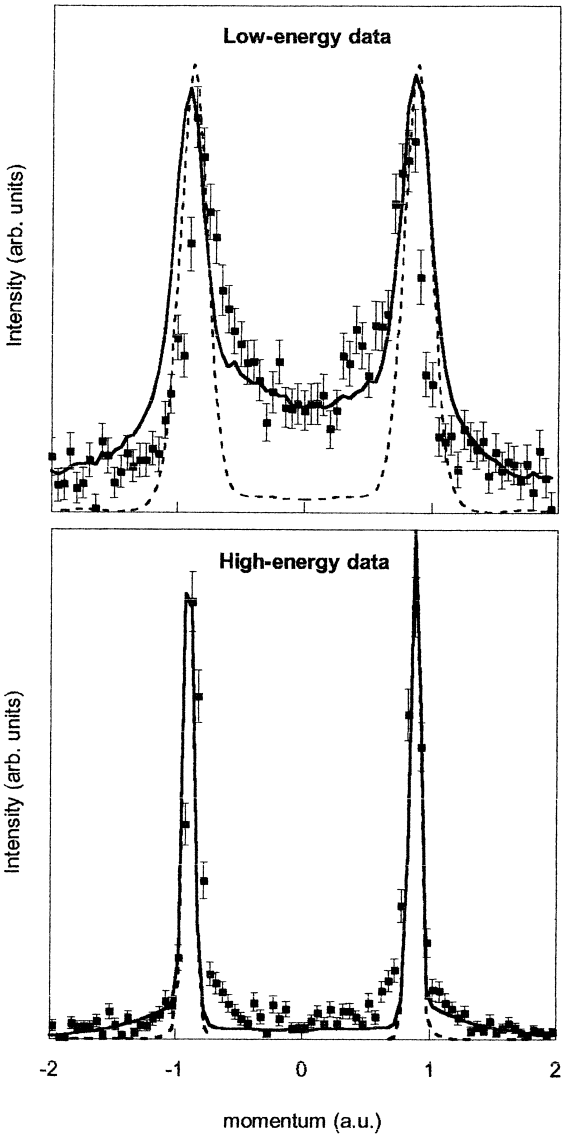

Fig. 4. The measured momentum density distribution near E_f. The top panel shows the data previously obtained using the Flinders University spectrometer (from Ref. [12]). The lower panel shows the current data. The dashed line is the result of the LMTO calculation corrected for finite energy and momentum resolution, whereas the full line is this theory corrected for multiple scattering.

reproduced by the LMTO calculation. We observe a peak value of 0.88 a.u. This difference is due to the sampling of electrons slightly away from the Fermi level due to finite energy resolution ($\lesssim$1.8 eV). Including the lattice potential in the calculation has only very minor effects on the theory and does not contribute to the broadening. It is washed out completely if the finite energy resolution is taken into account.

The observed momentum distribution consist of a main narrow component and a smaller, broader component. The width of the narrow component can be completely understood from the finite energy resolution, and therefore sets only an upper limit on the present momentum resolution ($\Delta p_y < 0.1$ a.u). Elastic scattering gives rise to a very small flat background under these sharp peaks as can be seen from the Monte Carlo simulations (solid line in Fig. 4. The origin of the broader component, most clearly visible at the small momentum side of the peaks is still under investigation.

In conclusion we have shown that we can measure energy-resolved momentum profiles of unsurpassed quality. Planned improvement in energy resolution and sample preparation (removal of the carbon layer by sputtering) should result in very clean measurements of the spectral function, bringing out the full potential of the EMS technique for studying the electronic structure of solids.

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