Condensed matter electron momentum spectrometer with parallel detection in energy and momentum

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An electron momentum spectrometer has been constructed which measures electron binding energies and momenta by fully determining the kinematics of the incident, scattered, and ejected electrons resulting from (e,2e) ionizing collisions in a thin solid foil. The spectrometer operates with incident beam energies of 20–30 keV in an asymmetric, non-coplanar scattering geometry. Bethe ridge kinematics are used which for 20 keV incident energy has scattered electron energies of 18.8 keV at a polar angle of \( \theta_e = 14^\circ \) and azimuthal angles \( \phi_e \) in the range from \(-18^\circ\) to \(+18^\circ\) and ejected electrons of 1.2 keV and \( \theta_e = 76^\circ \) with \( \phi_e = \pi / 2 \) with \( \phi_e \). The technique uses transmission through the target foil, but it is most sensitive to the surface from which the 1.2 keV electrons emerge, to a depth of about 2 nm. Scattered and ejected electron energies and azimuthal angles are detected in parallel using position sensitive detection, yielding true coincidence count rates of 6 Hz from a 5.5 nm thick evaporated carbon target and an incident beam current of around 100 nA. The energy resolution is approximately 1.3 eV and momentum resolution approximately 0.15 \( a_0^{-1} \). The energy resolution could readily be improved by monochromating the incident electron beam.

I. INTRODUCTION

The first successful electron momentum spectroscopy (EMS) experiments were reported over 20 years ago, involving studies of thin film targets, with energy resolution of about 64 eV and momentum resolutions of 1.5–2 \( a_0^{-1} \). With such low momentum and energy resolution no meaningful comparison with theory could be made in the valence region, although useful information was obtained for the core state. It was, however, applied to gas targets at about the same time with much improved energy and momentum resolution. This work on argon showed for the first time the importance of correlation effects in the inner valence region of atoms. Since then the technique of EMS has developed into a powerful tool for the study of target electronic structure in the gas phase via high energy, high momentum transfer (e,2e) reactions. EMS of solids has progressed much more slowly due to the difficulty of studying high density targets in transmission. The multiple scattering effects that occur in solids must be minimized by using a high energy incident electron beam, but as the incident beam energy is increased the (e,2e) cross section decreases and accurate definition of the different electron momenta becomes more difficult. Ritter et al. was the first group to resolve the valence structure of a solid target, being an amorphous carbon film, with 6 eV energy resolution and an incident beam energy of 25 keV. Since then two other groups have improved on these early measurements by using multidetection of the outgoing electron energies, a broad range of binding energies being observed at a single value of momentum. This paper describes a new spectrometer which uses multidetection of both the angles and energies of the outgoing electrons. For the first time a range of binding energies and recoil momenta are measured in parallel, leading to a greatly enhanced sensitivity of the method.

EMS is a conceptually straightforward way of directly measuring the electronic structure of materials. In (e,2e) spectroscopy a high energy incident electron knocks out an electron from the target system and the two outgoing electrons are subsequently detected in coincidence. Measuring the incident, \( k_i \), and outgoing, \( k_s \) and \( k_e \), momenta allows the target electron binding energy, \( \varepsilon \), and target recoil momentum, \( q \), to be determined using conservation of energy and momentum:

\[
\varepsilon = E_s + E_e - E_i, \tag{1}
\]

\[
q = k_i - k_s - k_e, \tag{2}
\]

where by convention the outgoing electron with the highest energy is referred to as the scattered electron and the electron with lower energy is called the ejected electron.

At high energy and momentum transfer \( K = |k_s - k_i| \), the \( (e,2e) \) collisions involve the clean knockout of the target electron (a so-called binary collision) and the recoil momentum is equal and opposite to the momentum of the struck target. It is well established that under these conditions, namely sufficiently high electron energies and momentum transfer, \( K \), the \( (e,2e) \) cross section is proportional to the probability of finding a target electron with a particular binding energy and momentum, multiplied by the half-off-shell Mott electron-electron scattering cross section. Under suitable conditions, the latter cross section has negligible variation over the range of momenta of interest. In the independent particle approximation the cross section is directly proportional to the square of the momentum space wavefunction of the electron. If electron-electron correlations are important in the final state then this has to be multiplied by the spectroscopic factor or pole strength, which is the probability

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that the final state contains the independent particle target one-hole state. If correlations are important in the initial state, the measured momentum densities will reflect this. For a solid the \((e,2e)\) cross section for ejection of electrons from a band is given by

\[
\sigma(q,e) = c \rho_\alpha(e^\alpha(q),q),
\]

where \(c\) depends on kinematical factors and is constant in the present experimental arrangement and \(\rho_\alpha\) is the one electron spectral momentum density of the target and \(e^\alpha(q)\) is the energy of band \(\alpha\). In the independent particle approximation, for a crystal

\[
\rho_\alpha(e^\alpha(q),q) = \sum_G |\phi_\alpha^G(k+G)|^2 \delta(q,k+G) \delta(e - e^\alpha(q)),
\]

where \(k\) is the crystal momentum, \(G\) is a reciprocal lattice vector and \(\phi_\alpha^G(q)\) the momentum space wavefunction of the target electron in band \(\alpha\). The detailed discussions of these points can be found in Refs. 4 and 5.

II. DESIGN CRITERIA

An electron momentum spectrometer requires a suitable electron beam source, a target, and a detection system for two electrons. The electron beam source provides a well collimated, parallel beam of electrons accelerated to a final energy \(E_f\) for ionization of the target. The detection system must provide energy, angle, and time resolved detection of scattered and ejected electrons resulting from ionization events. Time resolved detection allows the true coincidence rate to be determined by subtraction of random background events.

A. Scattering geometry

The design criteria for the spectrometer are of course high coincidence count rate, good energy resolution, and good momentum resolution. The scattering kinematics influences both the cross section and, hence cross rate, as well as the attainable energy and momentum resolution. EMS involves \((e,2e)\) collisions in which the ionized electron is knocked out cleanly, the so-called binary encounter region of the ionization process. This region of kinematics \(^9\) is called the Bethe ridge kinematics, for which it is easy to show that the scattering angle is given by

\[
\cos \theta = \left( k_s^2 + e \right) / k_s k_f.
\]

For asymmetric kinematics (in which \(k_s > k_f\)) the momentum transfer \(K\) is much smaller than for the normal symmetric kinematics for a given \(E_f\). This gives larger cross sections since the electron-electron collision cross section, which is included in the factor \(c\) in Eq. (3), is roughly proportional to \(K^{-4}\). \(K\) must however, be kept reasonably large, say \(K > 5 a_0^{-1}\), in order to ensure close electron-electron collisions and the validity of the binary encounter approximation. We chose \(\theta = 14^\circ\), which for an incident energy of 20 keV gives a "scattered" electron energy of 18.8 keV.

\[
K = 9.4 a_0^{-1}, E_f = 1.2 \text{ keV}, \text{ and } \theta = 76^\circ \text{ for } q = 0 \text{ [Eq. (2)].}
\]

Note that atomic units are used throughout, with \(\hbar = m_e = e = a_0 = 1\).

The criterion of good energy resolution can also best be met by the choice of an asymmetric scattering geometry since it allows one to overcome problems associated with drift in the high voltage power supply used to define the incident beam energy. Conventionally EMS has used symmetric geometry in which the outgoing electron energies are roughly half the incident beam energy. In gas phase experiments the incident beam energy is of the order of 1–2 keV, so it is straightforward to obtain energy stability of say 0.1 eV. In the present spectrometer the incident beam energy is typically 20 keV, with a maximum of 30 keV, defined by the potential applied to the electron gun filament, with the anode grounded. The best commercially available power supplies generally quote 50 ppm drift in the output voltage level over an 8 hour period, resulting in an electron beam energy drift of 1 eV over this time. By choosing asymmetric scattering kinematics in which the energy transfer to the target electron is of the order of 1 keV it is possible to use a single power supply to define both incident and scattered electron beam energies, as shown in Fig. 1. This means that drift in the incident beam energy is compensated by an equal drift in the scattered electron energy, while the effect of 1 eV drifts in both \(E_i\) and \(E_s\) on the conservation relations [Eqs. (1) and (2)] is negligible.

The energies of the incident and emitted electrons determine the extent to which multiple scattering influences the measured cross section. Such effects are discussed in detail elsewhere. \(^8,10\) With the choice of asymmetric kinematics and a lower energy "ejected" electron the spectrometer is more sensitive to the target region adjacent to the surface from which this electron emerges.

B. Momentum range

In order to maximize the count rate the spectrometer uses two dimensionless position sensitive detectors (PSDs).
Electron momentum spectrometer

III. VACUUM SYSTEM AND ELECTRON GUN

The vacuum system has been designed for UHV operation. Using sorption pumping and a 8 in. i.d. cryogenic pump to obtain pressures in the mid-10^-9 Torr range. This modest pressure is obtained prior to a bakeout of the system, and is reduced to the low 10^-10 Torr range after bakeout. A sample introduction system allows the targets to be changed without breaking the vacuum in the main chamber. The electron gun is an UHV compatible version of the design outlined by Schmoranzer et al., using a LaB6 cathode to minimize thermal energy broadening of the electron beam. The gun is mounted in an 8 in. nipple and can be isolated from the main chamber by a gate valve and is differentially pumped by a 20 /s ion pump. System alignment is achieved by the use of a collimating stage and Faraday cup mounted inside the main chamber to define the electron beam axis, and alignment apertures at the rear of the analyzers. A surveying telescope is used to ensure that the target is centered at the intersection of the lines of sight through the analyzers, which are prealigned with the collimator/Faraday cup mount. A set of Braun bek coils are aligned with the local magnetic field and used to reduce the field below 5 mG at the interaction region. The magnitude of the magnetic field over the scattering plane in the main chamber is below 30 mG.

IV. ELECTRON OPTICS

Electrostatic deflectors provide the energy analysis of the outgoing electrons. These are routinely capable of achieving better than 1% resolution of their mean pass energy. In order to achieve the overall (e,2e) energy resolution of about 1.3 eV the outgoing electrons are decelerated to about 100 eV before being dispersed in energy by the deflectors. Two different deflectors are used. These are a hemispherical deflector for the scattered electrons and a toroidal deflector for the ejected electrons. Each analyzer is designed to detect electrons over a pass band of about 20% of their mean pass energy using precalibrated position sensitive detectors. This allows parallel detection of a wide range of target electron binding energies. Each analyzer is also required to accept a range of azimuthal angles. It is the axial or near axial symmetry of each lens stack and deflector which allows parallel data acquisition over the desired ranges of azimuthal angles. This allows the spectrometer to simultaneously observe a large range of target electron momenta perpendicular to the incident beam (Fig. 2).

An overall sectional view of the electron analyzers is shown in Fig. 3, which shows the relative positions in the vacuum chamber, along with the Faraday cup and collimation stage for the incident beam.

A. Hemispherical analyzer

In the kinematics chosen for the experiment the scattered electron cone emerges to form an arc of radius r=d sin 14° at some distance, d, from the interaction region. Over a small range of azimuthal angles this arc is reasonably matched to the normal entrance conditions a hemispherical deflector of mean radius r, so that a hemispherical deflector can be used to analyse the scattered electrons (with fixed θ2), giving en-

Each detector collects electrons over a range of energies and azimuthal angles. The azimuthal angles accepted by the analyzers are restricted by slits at the entrance of each analyzer and limited by the location of the detector with respect to the symmetry axis. For the ejected electron analyzer the slits accept electrons with $-6^\circ < (φ_a - \pi) < 6^\circ$, of the maximum range that could be imaged onto the detector. The scattered electron analyzer has a maximum azimuthal range of $±18^\circ$, but was generally restricted to accept only $φ_a$ in the range $±10^\circ$ by adjusting the entrance slit. The azimuthal ranges determined the momentum range of the spectrometer. This range is illustrated in Fig. 2, which shows the allowed momentum conserved region [Eq. (2)] for all possible combinations of $φ_a$ and $φ_e$ and $E_1=20$ keV, $E_s=18.8$ keV, and $E_x=1.2$ keV. For the case where the hemispherical azimuthal range is $±18^\circ$ many of the extra momentum points that are accessible are at momenta above $q=2a_0^{-1}$. Since the $(e,2e)$ cross section is almost zero in these regions for valence electrons of say amorphous carbon the azimuthal range of the hemispherical analyzer was reduced to minimize the background count rate.

FIG. 2. Momentum range of the spectrometer for azimuthal acceptance angles in the range (a) $φ_a=±10^\circ$, $(φ_a-\pi)=±0^\circ$, and (b) $φ_a=±18^\circ$, $(φ_a-\pi)=±7^\circ$. The hatched regions show the momenta that are detected for all possible combinations of the angles $φ_a$ and $φ_e$, the momentum $q_x$ along the incident beam direction being set at zero.
energy dispersion, while maintaining the azimuthal angle information through the axial symmetry of the deflector. In practice some approximations need to be made in the implementation of this design; these will be discussed below.

The mean radius of the hemispherical deflector is 48.5 mm. This is determined by the requirement of obtaining an energy resolution of much better than 1 eV, assuming an analyzer pass energy of 100 eV, a lens magnification of unity, and an electron beam diameter at the target of 0.2 mm. At the same time the analyzer needed to be accommodated within a vacuum chamber of 600 mm diameter. Given these conditions and the fixed polar angle, the choice of mean radius of the deflector also influences the following properties of the analyzer; (1) energy resolution, (2) energy range, (3) azimuthal angle resolution, (4) azimuthal range, and (5) distance of the entrance plane from the interaction region.

The available azimuthal angle and energy ranges are also strongly influenced by the size of the PSD used to detect the electrons. By using a 40 mm active diameter PSD and a mean radius of 48.5 mm for the hemispherical analyzer (with inner radius 33.5 mm and outer radius 63.5 mm) the energy range that is imaged on the detector is 17% of the pass energy, and the azimuthal angular range available is ±18° (Fig. 7, to be discussed later).

The scattered electron trajectories are, however, not ideal for input to a hemispherical deflector. Ideally trajectories enter perpendicular to the entrance plane (in the plane containing the symmetry axis) within some small pencil angle.14 It can be seen that this condition of perpendicular entrance angles at \( \phi = 0 \) is satisfied by having the symmetry axis of the deflector tilted at 14° to the system symmetry axis given by \( k_\parallel \) (see Fig. 4).

In the present analyzer different azimuthal angles are accepted by matching the constant mean radius of the deflector with the arc of trajectories passing through the deflector entrance plane. The acceptance solid angle is given by the range of angles \( \theta_s = 14.0° \pm 0.1° \) and \( \phi_s = 18° \) to +18°, fixed by the geometry of the target and entrance slit.

To see the effect of increasing azimuthal angles, consider as an example the trajectory at \( \phi_s = 180° \) (Fig. 4). Clearly that trajectory would not be transmitted by the deflector, not only because it is not perpendicular to the entrance plane, but also because it intersects the entrance plane well away from the mean radius of the deflector. Thus the emerging trajectories are not ideal input trajectories for a hemispherical deflector. Furthermore, deceleration of the electrons by a lens system requires a lens which will accommodate the change in distance between the scattering center and the deflector entrance plane for changing \( \phi_s \). Three factors which need to be considered in implementing parallel detection of a range of azimuthal angles using a hemispherical deflector are: (1) the trajectories for \( \phi_s \neq 0 \) will no longer be perpendicular to the entrance plane of the deflector; (2) the distance from the target interaction region to the deflector entrance plane is not constant; and (3) the arc of azimuthal angles is not a circle in the entrance plane of the deflector.

Each of these points will be discussed in the following paragraphs. With reference to Fig. 4 these points are all obvious for the case \( \phi_s = 180° \). However analytic geometry considerations show that in all cases the effects are negligible over the required small range of azimuthal angles. This is further illustrated by the uniformity of the calibration image of Fig. 7, which will be discussed later.

Concerning point (1) above it is clear that the trajectory at \( \phi_s = 180° \) would not be transmitted by the deflector even if it entered at or near to the mean radius. It is not perpendicular to the entrance plane, with the deviation from perpendicular being \( 2\theta_s \). In practice the angle between a vector perpendicular to the entrance plane of the deflector and an incoming trajectory can be separated into two components, \( \alpha \)
Sectional view of the scattered electron cone intercepting the entrance plane of a hemispherical deflector, with the mean radius through the hemispheres shown. At $\phi_s = 180^\circ$ the scattered electron cone no longer intercepts at the mean radius of the analyzer, and the trajectories are no longer perpendicular to the entrance plane.

$\phi_s = 180^\circ$

$\phi_s = 0^\circ$

$\theta_s = \theta_s$

$R = 48.5 \text{ mm}$

$\phi_s = 20^\circ$

FIG. 4. Sectional view of the scattered electron cone intercepting the entrance plane of a hemispherical deflector, with the mean radius through the hemispheres shown. At $\phi_s = 180^\circ$ the scattered electron cone no longer intercepts at the mean radius of the analyzer, and the trajectories are no longer perpendicular to the entrance plane.

FIG. 5. Plot showing the ellipse (solid curve) generated by the intersection of the hemispherical deflector entrance plane with the scattered electron cone at $\theta_s = 14^\circ$. The hemispherical deflector mean radius of 48.5 mm is also shown (dashed curve).

and $\beta$. The component $\alpha$ lies in a radial plane, and the component $\beta$ is in the axial plane (perpendicular to the radial plane for a given $\phi_s$). The component $\beta$ has no effect on the imaging of the trajectory (neglecting fringing fields) because of the symmetry of the hemispherical deflector. Electron trajectories are influenced by the component $\alpha$, which affects the eccentricity of the electron orbit in the $1/r^2$ field of the deflector. If $\alpha$ is too large for a given deflector the electron will not be transmitted, rather colliding with the wall of the deflector. A general expression for $\alpha$ as a function of $\phi_s$ is given by

$$\alpha = \cos^{-1} \left[ \frac{\sin^2 \theta_s \cos \phi_s + \cos^2 \theta_s}{(\sin^2 \theta_s \cos^2 \phi_s + \cos^2 \theta_s)^{1/2}} \right].$$

From Eq. (6) the deviation $\alpha$ is below 1° for $\phi_s < 20^\circ$ and $\theta_s = 14^\circ$, which is less than the range of pencil angles entering the analyzer.

Concerning point (2) above, the distance from the collision region to the tilted entrance plane of the analyzer is given as a function of $\phi_s$ by

$$d_\phi = \frac{r}{\sin \theta_s (\cos \theta_s + \sin \theta_s \tan \theta_s \cos \phi_s)}.$$

The change in distance $d_\phi - d_0$ calculated for the analyzer, with $d_0 = 19.5 \text{ mm}$, is below 0.6 mm for $\phi_s < 20^\circ$.

The arc of intersection of the hemispherical deflector with the scattered electron cone at $14^\circ$ is an ellipse [point (3) above]. Figure 5 shows this ellipse for a choice of $r = 50 \text{ mm}$ and $\theta_s = 14^\circ$. Clearly, for $\phi_s < 20^\circ$ the ellipse and the mean radius are well matched. The difference between the two curves is less than 0.5 mm over this range of azimuthal angles.

The suitability of these trajectories for input to a hemispherical deflector is therefore demonstrated, provided that the small deviations from ideal input can be accommodated. We now discuss the design of a deceleration lens for coupling to the deflector. This lens is required to decelerate from nominally 18.8 keV to 100 eV, while maintaining the azimuthal angle information and providing a focus within criteria required for optimal energy resolution of the hemispherical deflector. Design of the lens is considerably simplified by the fixed kinematics of the experiment which means that in any given experiment the analysis energy is fixed. Furthermore, a momentum resolution of 0.1 $a_0^{-1}$ requires the range of accepted polar angles to be restricted to $14^\circ \pm 0.1^\circ$.

The image of the incident beam spot at the target is used as the entrance window of the deceleration lens that precedes the hemispherical deflector. The deceleration lens images the outgoing electrons at the entrance to the deflector at a kinetic energy of nominally 100 eV to obtain the desired energy resolution. The pupil is defined by the slit at the analyzer entrance, which controls the range of polar angles accepted by the lens. The electron trajectories enter the lens stack with zero beam angle and a pencil angle of 0.2°. The desired linear and angular magnification provided by this lens are then determined using the criteria outlined by Kuyatt and Simpson for obtaining a particular energy resolution from a hemispherical deflector.

The deceleration lens is in the form of curved slits which are open over the range of required azimuthal angles—a conical slit lens. Conical slit lenses have been used before as input and output optics for analyzers, but no focal lengths were given and the lens design was by empirical modeling. For the present spectrometer a version of the SLAC electron trajectory program is used to model the electron optics. Figure 6 shows a sectional view of the lens for $\phi_s = 0^\circ$. The lens has rotational symmetry about the horizontal centerline given by the incident beam direction. Trajectories shown in Fig. 6 are generated by the SLAC optics program. Scattered electrons are decelerated to about 100 eV by this lens, with a typical deceleration of around 200:1 and a filling factor of the lens of below 30%.

An initial deceleration stage is formed by the first and second lens elements. The first lens element is at ground potential. Typically the second lens was operated at $-17.888 \text{ kV}$, allowing the azimuthal angle information to be retained.


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These lens elements were operated at potentials of about 19.7 kV, to decelerate to nominally 100 eV. The hatched regions represent the analyzer entrance plane from 195 to 195.6 mm as the azimuthal angle increases to $\phi = 20^\circ$. This increase in the trajectory path length is accommodated by machining the final stage lens about the same symmetry axis as the hemispherical deflector. In this way the distance between the penultimate and final deceleration lens increases with increasing azimuthal angle, since the thickness of lens element five changes. To minimize aberrations due to this the output of the field lens is chosen to be imaged to infinity. This means that the object of the final stage lens is at the same apparent position, regardless of the azimuthal angle in the lens. The lens is not modeled for its effect on the azimuthal range and focusing. In practice the final stage lens can be seen to change the apparent angular range. As the deceleration ratio of the final lens is increased the available azimuthal angular range is reduced. This effect occurs because for non-zero azimuthal angles the trajectories are not perpendicular to the plane of the lens and hence diverge upon deceleration.

Materials used in the construction of the hemispherical analyzer are aluminium, machinable ceramic, molybdenum, and sapphire ball insulators. At the exit to the hemispherical deflectors and in front of the PSD there is a 90% transmission stainless steel mesh (50 lines per inch, 0.001 in. wire) which defines the potential at the exit to be at the mean pass energy. Electron detection after the exit of the analyzer is by a 40 mm active area position sensitive detector, consisting of an arrangement of microchannel plates followed by a resistive anode (Quantar Technology Inc. Model 3395A MCP/RAE detector). Signals from this detector are processed by electronics to be discussed later.

Edge effects by the lens on the different azimuthal angles were avoided by machining the slits over a much wider azimuthal range than required by the spectrometer, always ending at least three times the lens gap away from any electron trajectories. There is no degradation of the quality of the analyzer resolution across the azimuthal angular range. This is clearly illustrated in Fig. 7 which shows a calibration image at the exit of the detector.
B. Calibration of hemispherical analyzer

Calibration of the analyzer is carried out using a set of apertures mounted on the front of the lens stack and elastic scattering measurements. Electrons leaving the target must pass through the calibration apertures before entering the optics. Because the space between these apertures and the target is field free there are no angular distortions of the incoming trajectories.

Elastic scattering from a thin (5–10 nm) self-supporting amorphous carbon foil provides a precise energy and angle calibration of the detector. A series of peaks are collected corresponding to known electron energies and angles. Figure 7 shows typical calibration data with the intensity scale shown in gray scale. This was collected by incrementing the incident beam energy in 2 eV steps from 18.780 to 18.794 keV, with the analyzer set to collect 18.788 keV electrons on the mean radius of the hemispherical deflector exit at a pass energy of 100 eV. The calibration apertures at the front of the lens system were 0.2 mm diameter holes drilled into an 0.1 mm thick molybdenum foil at \( \phi_0 = 0 \), \( \pm 3^\circ \), \( \pm 7^\circ \), \( \pm 11^\circ \), and \( \pm 15^\circ \).

The data of Fig. 7 is used as the input to a series of programs. A centroid for the \((x,y)\) position of each peak is automatically located by peak recognition software and tabulated along with the known energy and angle of each peak. A pair of two dimensional fourth order polynomials are fitted to the centroid data using a least squares method. These polynomials are subsequently used to define the transformation between the \((x,y)\) arrival position coordinate (as determined by charge division) and the energy and angle of each detected electron.

The quality of this fitting procedure was illustrated by the rms and maximum deviations of the resulting polynomials from the centroid data. This was typically 0.1 and 0.2 eV, respectively, for the energy fit for 100 eV pass energy and 0.1° and 0.3° for the angular fit. The values for the rms deviation of the polynomials from the centroids are well within the resolution of the electron optics.

C. Ejected electron analyzer

In the chosen kinematics the scattered and ejected electrons travel in approximately orthogonal directions since the momentum of the ejected electron is roughly equal to the momentum transfer. Thus the ejected electrons emerge from the target with a polar angle of \( \theta_e = 76^\circ \) (for \( q=0 \), \( e=0 \) and neglecting relativistic effects). At such a large polar angle the ejected electrons are emerging from the interaction region traveling radially, perpendicular to the symmetry axis. Tofoletto et al., following the notation of Wollnik, present transfer matrices for polar trajectories through toroidal deflectors. The deflector was chosen with the ratio between the minor radius, \( a \), and major radius, \( b \), as \( c=a/b = 0.8 \) in order to optimize its angular and energy focusing.

The analyzer size was chosen such that \( a=80 \) mm, \( b=100 \) mm. This made possible a spacing of 30 mm between the target and the 0.25 mm wide lens entrance slit, necessary for limiting the polar acceptance angle to 0.5° with the given target area. A distance of 50 mm was then available for the deceleration lens. Momentum resolution requirements constrain the range of polar angles entering the analyzer to be \( 76^\circ \pm 0.25^\circ \).

Design of the deceleration lens proceeded along the same lines as for the hemispherical analyzer lens. The toroidal deflectors are truncated by parallel plates at 75 mm above and below the spectrometer plane since the toroidal analyzer is only required to accept a small range of azimuthal angles. These endplates are biased at the pass voltage of the analyzer. The lens was again empirically designed using the SLAC electron trajectory program. Results from this program for the lens are shown in Fig. 8. This lens is a cylindrical slit lens, with deceleration and focusing for trajectories traveling radially, perpendicular to the symmetry axis. Typical voltages on the lens elements are \( V_1 = 0 \), \( V_2 = -776 \), \( V_3 = V_4 = -861 \), \( V_4 = -1069 \), and \( V_6 = -1092 \) V.

![Computer simulation of the electron trajectories in the toroidal analyzer deceleration lens, showing the 1.2 keV ejected electrons decelerated to 100 eV and focused at the entrance to the toroidal deflectors. Typical voltages are \( V_1 = 0 \), \( V_2 = -776 \), \( V_3 = V_4 = -861 \), \( V_4 = -1069 \), and \( V_6 = -1092 \) V.](image)

FIG. 8. Computer simulation of the electron trajectories in the toroidal analyzer deceleration lens, showing the 1.2 keV ejected electrons decelerated to 100 eV and focused at the entrance to the toroidal deflectors. Typical voltages are \( V_1 = 0 \), \( V_2 = -776 \), \( V_3 = V_4 = -861 \), \( V_4 = -1069 \), and \( V_6 = -1092 \) V.
$V_3 = V_5 = -861$, $V_4 = -1069$, and $V_6 = -1092$ V, respectively. The operating principles for this lens are the same as for the hemispherical analyzer, with an initial deceleration stage, followed by a field lens and final imaging lens.

Imaging of the electrons at the exit of the deflector is by a 40 mm active diameter PSD. The potential at the exit of the analyzer is defined by an 80% transmission stainless steel mesh (100 lines per inch, 0.001 in. wire) that is spot welded to a molybdenum plate and mounted to the front of the PSD. This assembly is then mounted in an aluminium enclosure. A conical surface is machined on the face of this enclosure where it attaches to the toroidal deflector exit, to match the sector angle of 144°. The deflector itself is ended at a sector angle of 141°, with the detector set at 144° to give an approximate Herzog correction$^{22}$ to the field at the deflector exit, with the exit potential set at the mid-deflector potential.

D. Calibration of toroidal analyzer

The calibration of the toroidal analyzer is performed using the technique described earlier for the hemispherical analyzer, resulting in a calibration image similar to Fig. 7. Electrons enter the analyzer via 0.16 mm diameter apertures machined at $\phi_e = 0, \pm 2.55^\circ, \pm 5.1^\circ$, and $\pm 7.7^\circ$. For the toroidal analyzer the calibration grid that results with these apertures has five points across the detector in the azimuthal direction, for most of the energy range. There are typically 12 columns of peaks across the plate in the energy direction, resulting from moving the elastic peak across the plate in 4 eV steps from 1172 to 1212 eV, with the pass energy set at 200 eV. A slice across each detector calibration image in the azimuthal direction is shown in Fig. 9 for each analyzer, which clearly illustrates the excellent azimuthal angular resolution. Differences in the peak heights are due to variations in the detector efficiency. The even shape and spacing of these peaks with respect to angle shows that distortions introduced by the electron optics are negligible.

V. ELECTRONICS

A. Position sensitive detectors and fast timing circuits

The PSDs at the exit of the analyzers are microchannel plate and resistive anode encoder combinations MCP/RAE (Quantar Technology Model 3395A Sensor). PSDs of this type have been used in electron momentum spectroscopy experiments as one dimensional detectors.$^{23}$ This is the first time they have been used for parallel detection in both energy and angle for electron momentum spectroscopy, which combines fast coincidence techniques with parallel two dimensional position determination in two detectors. Position determination is by charge division: the total charge collected by the Gear RAE$^{24}$ diffuses to the four corners, with the quantity arriving at each corner depending on the arrival position of the charge pulse due to the relative resistances and capacitances between the arrival position and the four corners collecting the charge. Electronic output from the analyzers is in the form of five pulses from each detector. Four pulses are for the position determination and one pulse is for coincidence timing.

The four position charge pulses from the four corners of the resistive anode are capacitatively decoupled from the anode voltage and input to preamplifiers (Ortec 142A), then the signals are amplified and shaped by Ortec 855 spectroscopy amplifiers. The spectroscopy amplifier produces a bipolar Gaussian shaped output pulse with a shaping time constant of 0.5 μs. The circuitry for the position pulses from each detector is shown schematically in Fig. 10. The fast timing pulses are taken from the exit of the microchannel plate assembly, and are decoupled from the voltage at the rear of the channel plate stack by pulse transformers as shown in Fig. 11.

In the hemispherical analyzer the pulse transformer is floating at about −16 kV and used for pulse inversion, with decoupling from the high voltage by capacitors (HEC, 340 pF, 40 kV). For each charge cloud leaving the rear channel-plate a fast timing pulse is produced. These pulses are preamplified (Ortec, VT120) and input to a constant fraction discriminator (CFD; Tennellec, TC454). Following the preamplifier the negative going pulses typically have 2 ns rise-times with amplitudes between 200 and 600 mV. The CFD serves to minimize timing jitter due to the variation in pulse amplitude and to reject noise and reflections of the primary pulses below the level of a discrimination threshold. The exact set-up of the signal decoupling for the fast timing
pulses is determined empirically since it is sensitive to the positioning of the various components.

Fast logic pulses from the CFDs are used as start and stop input for the time to amplitude converter (TAC; Ortec 567). In coincidence mode the start pulse is taken from the hemispherical analyzer and the stop pulse from the toroidal analyzer after a nominal 20 ns delay. The resulting timing spectrum shows a peak due to true coincidences, superimposed on a flat background. The TAC is also gated by pulses detected by a noise antenna circuit (Fig. 11) so that TAC output is inhibited when spurious pulses are detected.

**B. Multiparameter acquisition system**

Position and TAC output pulses are fed to a multiparameter acquisition system (MAS) which forms the interface between the NIM electronics and the computer. The MAS was constructed in-house and comprised ten channels of parallel data acquisition. The timing signal from the TAC and the eight amplified charge pulses from the corners of the two PSDs are fed into nine gated 12 bit analog to digital converters with a conversion time of 15 μs. Pulse pile-up rejection was set at 70 μs. A dual channel buffer memory system allows data conversion and subsequent storage in one 8k
buffer, while the second 8k buffer is being read by the online computer. The tenth input channel is in digital form directly from the separation voltage power supply controller.

The nine analog signals are gated into the MAS by a strobe pulse from the TAC, which indicates that a true time conversion has occurred, the TAC being set to give a conversion over a range of 200 ns between start and stop pulses. For each strobe pulse a data set (ten channels) is stored in the buffer. This method of gating is a crucial feature of the data acquisition system, since data processing is only required when a correlated pair of events (within the 200 ns TAC span) is observed. This means that most of the single electron counts detected by each analyzer are rejected. Since the single electron count rate is around 40 kHz (limited by pulse pileup) compared to the TAC strobe rate of around 200 Hz, the data storage and processing requirements are greatly reduced. Under software control the stored data is periodically transferred to the computer for processing.

The base width of the coincidence peak from the TAC is about 20 ns. Different electron flight times in the analyzers are the dominant contributions to the base width of the timing peak. This effect has been described in detail for hemispherical analyzers and results from the different path lengths and velocities of the electrons being detected. As a result the signal to noise ratio of the timing information is lowered, by spreading the true coincidence data over a larger number of channels. Lower and Weigold have detailed a method which reduces the width of the timing peak by applying an analyzer transit time correction to each incoming timing pulse according to the correlated electron energy. This method involved the electronic manipulation of the timing pulses according to the arrival positions. In the current experiments the data correction is performed by software after the timing spectrum from the TAC is input to the computer via the MAS. Figure 12 shows an uncorrected timing spectrum, and timing spectra with the flight time correction applied for each analyzer independently and for both analyzers together. The improvement in the timing for the true coincidences is clearly seen. Also shown is the effect on the accidental “noise” coincidences due to signal not originating for true electrons. The real coincidence peak appears after the noise signal since the mean electron flight time in the toroidal analyzer is about 15 ns longer than for the electrons in the hemispherical analyzer, and the signal from the toroidal analyzer is used to stop the TAC. In contrast to the true signal from electrons, the noise spikes are spread out by the timing correction. During normal operation care is taken to eliminate these spurious noise peaks.

The computer used for reading and analysis of the data from the MAS is a Compaq Deskpro 486/25, operating in protected mode using a DOS extender. The analysis software (Watcom C programming language) initially checks that all pulses fall within defined upper and lower thresholds, to avoid processing noise and/or pulses which had saturated in the amplification stage. This threshold requirement, in combination with the gating of the MAS by the fast timing circuitry and the different frequency responses of the fast timing and slow position encoding circuits, means that the system is essentially immune to noise pickup. All of the data for a given experiment is stored in binary as a list of 16 bit binary vectors. Each vector contains six numbers that remain after processing the charge pulse data from the PSDs into (x,y) coordinates by charge division. These six numbers are then the (x,y) coordinates for the two PSDs, the TAC output, and the separation energy from the gun power supply. For a given experiment this is in general a large quantity of data. Data are therefore transferred periodically to tape for storage in blocks of the order of 10's of megabytes, to clear the 160 megabyte hard drive of the computer for further data acquisition. This raw data can be reanalysed after the experiment if for example the calibration of one of the detectors is accidentally changed. (Note that in general the calibration of the analyzers is quite stable.) The data is also processed online to produce a histogram corresponding to the spectral momentum density of the target material. In addition to this histogram the software allows the incoming data to be displayed in a number of diagnostic plots, including the timing spectra of Fig. 11, individual analyzer pulse height distributions separately from the four corners and summed, detector images, angular and energy distributions and various other diagnostic information to allow the operator to ensure that the spectrometer is operating correctly.

Eight of the channels read in from the MAS are the two sets of four pulses from the four corners of the resistive anode encoders of the PSDs. These are converted to two sets of (x,y) coordinates for each detector. The electron energies and angles are then calculated using these (x,y) coordinates as input to conversion relations determined by the pre-measured calibration information for each detector. Complete details of the software algorithms developed for this calibration are given by Caprari. The electron energies are used to determine the flight time correction to the timing spectrum from the TAC (channel nine of the MAS). The tenth channel is used to input the separation energy from the programmable power supply controller.

The binding energy and momentum of the struck elec-
tron is then calculated using the energy and angle information from each detector and the current energy increment of the incident beam according to the separation voltage power supply controller. The full range of target electron momenta is collected in parallel. The range of binding energies collected in parallel by the analyzers is scanned over a wider range of binding energies by the separation voltage power supply, under computer control, in a binning mode similar to that described by Cook et al. 23

The true coincidence counts, $N_i$, are determined in the usual way, according to the timing information. 4 For each energy and momentum bin the data correlated with the background region of the timing spectrum, $N_b$, is subtracted from the data correlated with the coincidence signal region, $N_c$, after dividing the background counts by the background to coincidence window ratio, $r$. That is,

$$N_i = N_c - N_b/r .$$

The statistical error associated with $N_i$ is

$$\Delta N_i = (N_c + N_b/r^2)^{1/2} .$$

A correction for the instrumental response function is applied to each energy-momentum bin, by normalizing each bin to the total number of background counts in that bin. This correction compensates for the variations in detection efficiency of the detectors and for the variations in the momentum range illustrated in Fig. 2. For isotropic amorphous targets any systematic offset in the azimuthal angle sum ($\phi_1 + \phi_2$) is identified in the software and subtracted in the analysis of the data. This, as well as a detailed description of the error analysis is given in Ref. 76. The effect of variations in detector efficiency on a broad range of the energy dimension is compensated for by the binning mode of taking data. 23

VI. RESULTS

The energy and momentum resolution of the spectrometer is limited by the energy and angular resolutions of the incident beam and the two analyzers. In a gas (e,2e) experiment the coincidence energy resolution may be easily measured since there are targets with well separated bound states, for example the 3p−1 transition in argon, so an attempt was made to determine the energy resolution of the spectrometer using argon as a target gas. The width of the main Gaussian in a fit of two Gaussian peaks to the resulting argon data was 2 eV, a rather poorer energy resolution than expected for the spectrometer. However for a gas target two effects seriously degrade the coincidence energy resolution. These are the effects of space charge in the initial stages of the electron gun, which significantly broadens the energy width of the incident beam, and the much greater length of the interaction region, which leads to angular and hence energy broadening. The gas phase energy resolution can therefore only be an upper limit to that observed with a solid target. The higher total electron beam current used for the gas phase experiment (about 10 μA compared to about 400 nA in the solid state experiments) results in significant space charge contributions to the incident beam energy spread. Using low beam current (<100 nA) the elastic scattering energy width is 0.6 and 0.7 eV for the solid and gas targets, respectively. For beam currents greater than 0.5 μA at the interaction region the gas elastic scattering energy width broadens to around 2 eV. The actual value is strongly dependent on the focusing conditions of the electron gun, particularly since an increasing fraction of the total current from the gun is skimmed off by the 0.25 mm diameter collimating aperture before the interaction region, as the beam current is increased. To obtain 0.5 μA at the interaction region requires a total current from the gun of about 10 μA. No results were obtained for the elastic energy width in solid scattering at high beam currents due to limitations on the single electron count rate tolerated by the detectors.

Therefore in order to determine both the energy resolution and an absolute energy scale, the 1s−1 carbon core state was studied in an (e,2e) coincidence experiment on a 4.5 nm film, under the same conditions as the measurement of the valence spectral momentum density for this sample. The result is shown in Fig. 13. The count rate for this experiment was about 20 times lower than the rate for the valence electronic structure measurements. Although the quality of the data is rather poor, a narrow peak at 284.4 eV is clearly evident. The effects of multiple scattering, mainly due to plasmon excitation, can also be seen as a broad peak at 308 eV. A good fit to the core state is obtained using three Gaussians. The main Gaussian has full width at half-maximum of 2 eV at a binding energy of 284.4 eV. The other two Gaussians have widths of 3.2 and 14 eV at energies of 286.8 and 288.5 eV, respectively. These widths include the natural linewidth contribution that has been measured to be 1.54 eV for amorphous carbon using x-ray photoelectron spectroscopy with a resolution of 0.2 eV.22 Using this value for the natural linewidth means that the spectrometer has a resolution function described by a main Gaussian of width 1.3 eV, since the natural linewidth convoluted with this Gaussian yields the width of the fitted peak. The detailed
shape of the core hole state may depend on the different bonding environments of the carbon atoms in the amorphous carbon target.

Momentum resolution is controlled by the angular resolution of the incident, scattered, and ejected beams. In these experiments the incident electron beam divergence at the interaction region is neglected, with the momentum resolution dominated by the angular resolution of the analyzers, as determined by the size of the collision region, the geometry, and the electron optics. For polar angles the resolution, $\Delta \theta_a$, is controlled by the width of physical slits at the entrance of each analyzer, $w_a$, the distance of these slits from the interaction region, $d_s$, and the dimensions of the incident beam spot on the target as seen by the analyzer, $w_t$, according to

$$\Delta \theta_a = \frac{w_a + w_t}{d_s}. \quad (10)$$

Since there are no optics between the target and the entrance slits the polar angular resolution is controlled entirely by these geometrical factors. Note that the angle of the target with respect to the incident beam and the emerging beams will change the dimension of the beam spot as seen by each analyzer. In the present work the target foil is set up at 45° to the incident beam direction.

A wide range of azimuthal angles is accepted by each analyzer, so different azimuthal angles are resolved at the detector, with some contribution from the electron optics. An estimation of the azimuthal angular resolution can be made from the angular calibration data of Fig. 9. These plots show the series of peaks that result from electrons entering the analyzer through the calibration apertures.

The azimuthal angular resolution of each analyzer, $\Delta \phi_a$, is related to the peak widths according to

$$\Delta \phi_a = \Delta \phi_{m} - w_{ap}/d,$$  \quad (11)$$

where $\Delta \phi_{m}$ is the width of the peak at the detector and $w_{ap}/d$ gives the contribution due to angular width of the apertures. In the limit $w_{ap} \to 0$ the value $\Delta \phi_a$ gives the azimuthal resolution of the analyzer, since the different angles accepted by the calibration apertures will be resolved at the detector, as long as the PSD resolution is high enough.

In a gas experiment on argon the momentum profile of the 3p ground state was measured (Fig. 14). Because this state has zero cross section at $q=0$, it provides a good measure of the momentum resolution of the spectrometer. In practice the value obtained by this measurement is an angular average of the resolution function. Also it gives only an upper limit of the width of the momentum resolution function, since the interaction volume defined by the intersection of the gas beam with the electron beam is much larger than the solid target interaction volume because of the expansion of the gas upon leaving the 0.2 mm nozzle. With a gas target the interaction volume length is estimated to be 0.5 mm, with a diameter of 0.25 mm (as determined by the dimensions of the electron beam). These values, which lead to a total momentum resolution of $\Delta q=0.25a_0^{-1}$, were used to provide corrected theoretical argon 3p$^{-1}$ and 3s$^{-1}$ momentum profiles. The expected agreement between the PWIA calculation and the experimental results is shown in Fig. 14. Allowing for the different interaction volumes for gas and solid targets, we estimate from these measurements that the momentum resolution for solid state targets to be approximately $0.15a_0^{-1}$, in good agreement with the design parameter.

Typical true data coincidence count rates of 6 Hz with signal to background ratio of 1 are obtained with the spectrometer for a 5.5 nm thick amorphous carbon target. This is for an incident beam current of 150 nA and single electron count rates of 12 and 22 kHz for the hemispherical and toroidal analyzers, respectively.

A set of data that required about 100 h of data acquisition under these conditions is shown in Fig. 15. This data shows the spectral momentum density of a nominally 5.5 nm thick evaporated carbon target, with some contribution due to multiple scattering in the target. The dominant inelastic multiple scattering loss in amorphous carbon involves plasmon excitation, which shows a broad peak at 25 eV energy loss. Corrections for multiple scattering are straightforward and will be discussed in a forthcoming publication.26

The features at binding energies of 8 and 20 eV at zero momentum are associated with the outer and inner valence bands of amorphous carbon. Evaporated carbon is considered to consist of largely trigonally bonded carbon structures, showing graphitic nature. The EMS measurements in Fig. 15 roughly agree with the graphitic nature of evaporated carbon. The...
inner valence band correlates well with an angular average of the inner valence $\sigma$ band of graphite at low momenta. The momentum density for this band, which shows considerable dispersion, peaks at $q=0$. The outer valence band is at the same energy as the high momentum density regions of the $\pi$ band of graphite. The momentum density for this band peaks at about $1 e_{\text{F}}^{-1}$. These as well as other results are discussed in detail in Ref. 28.

VII. DISCUSSION

Clearly the technique of EMS is capable of revealing the electronic structure of thin films, providing information about the binding energy and occupation of states at different momenta. Some limitations are imposed by the difficulty in obtaining thin target materials as required by the transmission geometry. Nevertheless there is a significant amount of work to be done in characterizing targets, including the evaporated carbon target shown here and a form of diamond-like amorphous carbon. Since the spectrometer requires an incident beam current of the order of a hundred nanoamperes for these experiments the incident beam could be monochromated to provide improved energy resolution.

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