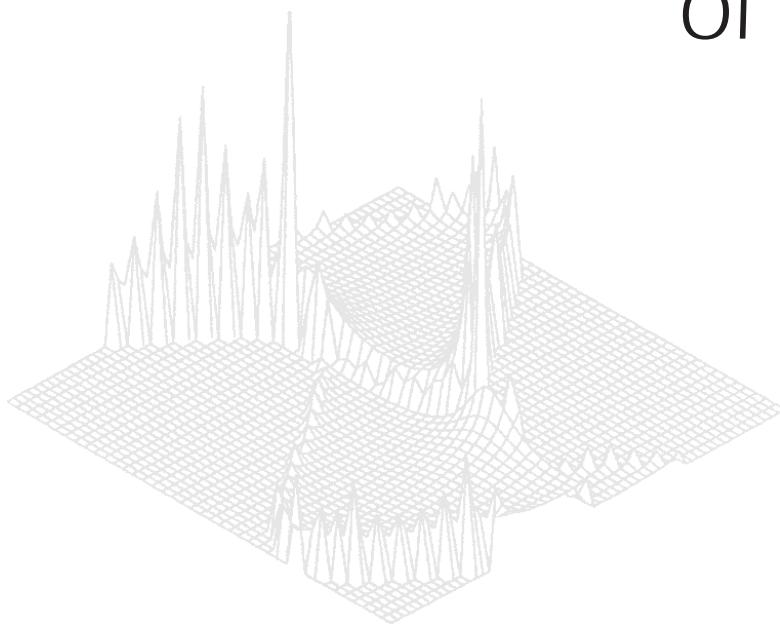

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Future Directions in Electron Momentum Spectroscopy of Matter*

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Abstract

The development of coincidence spectrometers with multivariable detection techniques, higher energy kinematics, monochromated and spin-polarised electron sources, will usher in a new generation of electron momentum spectroscopy revealing new electronic phenomena in atoms, molecules and solids. This will be enhanced by developments in target preparation, such as spin polarised, oriented and aligned atoms and molecules, radicals, surfaces and strongly correlated systems in condensed matter.

1. Introduction

Electron momentum spectroscopy (EMS) has been developed into a powerful tool for investigating the electronic structure of atoms, molecules and condensed matter. It provides unique information on the motion and correlation of the electrons in the sample of interest, particularly the valence electrons. It is especially sensitive to the low momentum components of the wavefunction, which in molecules is the chemically important region. It has established itself as a probe of unprecedented scope and detail for the single-particle (orbital) as well as correlated electronic structures of atoms and molecules. Recently it has been successfully applied to reveal details of spectral-momentum densities of electron bands in solids and surfaces. The application of EMS to structure studies is expanding quite rapidly, as demonstrated by the fact that it is currently being carried out in at least eleven centres in different parts of the world.

EMS is based on the high energy, kinematically complete, electron impact ionisation process, or $(e, 2e)$ collision. At high energies and high momentum transfers, the $(e, 2e)$ cross section takes a particularly simple form, being proportional to the square of the overlap between the initial target state and the final ion state wavefunctions. In the independent particle model for the target state, one obtains cross sections for transitions to final ion states which are proportional to the absolute square of the momentum space single particle orbital. The proportionality factor, known as the spectroscopic factor or pole strength, is the probability that the final ion state contains the one-hole configuration of the orbital. More generally, electron correlations in the initial state lead to changes

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in the momentum densities from that given by the simple independent particle model, and can lead to the transitions forbidden by that model.

Future applications of EMS are limited only by the ingenuity of the experimenter. They will require EMS spectrometers of greater sensitivity. This has already occurred to a large extent with the advent of the Flinders high energy asymmetric spectrometer designed for measurements of thin self-supporting solid samples in transmission geometries. This spectrometer incorporates two dimensional detectors capable of measuring simultaneously over a range of separation energies ω and a range of momenta \mathbf{k} . Similar multivariable detection techniques still need to be developed for gaseous targets, but plans to do so are well underway.

After outlining the theory of EMS, the future prospects for gaseous and solid targets will be discussed.

2. Theoretical Background

At high energies and in high momentum transfer collisions by the incident electron, the binary-encounter approximation is generally valid, and the cross section depends only on the coordinates and spins of the scattered and ejected electrons. Exchange terms between bound and free electrons are neglected. Such terms are negligible at high energies, since they involve the overlap of the wavefunctions of a free and bound electron, which is negligible at high momentum, particularly for valence electrons.

For multi-nucleus targets the total wavefunction describes the nuclear motion as well as the electronic states. These can be treated in the Born–Oppenheimer approximation. The states of nuclear motion (vibration and rotation) are usually not resolved and are summed over. The final states of nuclear motion can then be eliminated by closure, leaving only the appropriate initial-state averages. The vibrational average is described accurately by calculating the electronic states at the equilibrium positions of the nuclei (Dey *et al.* 1975). For unpolarised targets the average or sum over initial magnetic substate degeneracies of electronic motion in the case of atoms, the average over initial rotational states in the case of molecules, and the average over random orientations for polycrystalline solids, all amount to a spherical average, or an average over the direction of the electron momentum \mathbf{k} given by momentum conservation

$$\mathbf{k} = \mathbf{k}_f + \mathbf{k}_s - \mathbf{k}_0. \quad (1)$$

Here the subscripts 0, f and s denote respectively the incident electron and fast and slow emitted electrons. The separation or binding energy of the ejected electron is given by

$$\omega = E_0 - E_f - E_s. \quad (2)$$

In the plane wave impulse approximation, performing the sum and average over the magnetic substates of the external electrons, the differential cross section becomes (McCarthy and Weigold 1976)

$$\sigma_i(\mathbf{k}_f, \mathbf{k}_s, \mathbf{k}_0; \omega) = (2\pi)^4 \frac{k_f k_s}{k_0} f_{ee} \frac{N}{4\pi} \int d\hat{\mathbf{k}} |\langle i | a(\mathbf{k}, \omega) | t \rangle|^2 \delta(\omega - E_0 + E_f + E_s), \quad (3)$$

where $a(\mathbf{k}, \omega)$ is the annihilation operator which annihilates an electron of momentum \mathbf{k} and energy ω in the target electronic state t leaving the ion in electronic state i . Usually t is the ground state, but it can be an excited state (Zheng *et al.* 1990).

In (3) N is the electron multiplicity for electronic states defined for a fixed orientation to an arbitrary set of space-fixed axes, for radial states in the case of an atomic target. The two-electron collision factor f_{ee} is essentially the half-off-shell Mott electron-electron scattering cross section, and can be easily calculated. It is dominated by the momentum transfer as $|\mathbf{k}_0 - \mathbf{k}_f|^{-4}$. For EMS the kinematic conditions are chosen so that f_{ee} , k_0 , k_f , and k_s are very nearly constant. Therefore the cross section (3) depends only on \mathbf{k} and ω and is proportional to the energy-momentum spectral function

$$A(\mathbf{k}, \omega) = |\langle i | a(\mathbf{k}, \omega) | t \rangle|^2 \delta(\omega - \omega_i). \quad (4)$$

The spectral function is a one-electron function. We can introduce the concept of a normalised Dyson orbital α through (McCarthy and Weigold 1991)

$$\langle i | a(\mathbf{k}) | t \rangle = (S_i^\alpha)^{\frac{1}{2}} \phi_\alpha(\mathbf{k}), \quad (5)$$

which assigns certain final states i to an orbital manifold α . Here S_i^α is the spectroscopic factor or pole strength, which is the probability of the final state i containing the orbital α . If the target can be well described by the independent particle approximation, the orbital $\phi_\alpha(\mathbf{k})$ is the independent particle orbital (e.g. Hartree-Fock orbital).

For polarised or oriented atomic targets (Dorn *et al.* 1998; Zheng *et al.* 1990), for molecular oriented targets or for single crystal targets (Vos *et al.* 1997), the spherical averaging in (3) is no longer applicable. Thus for crystals one can measure the spectral momentum density along different cuts in momentum space. Similarly the use of incident polarised electrons modifies the initial magnetic substate average of the free electrons (Guo *et al.* 1996).

The spectral density $A(\mathbf{k}, \omega)$ contains much more information than just the band (or orbital) position in energy. The magnitude of $A(\mathbf{k}, \omega)$ is the probability of the electron (or more generally quasi-particle) having momentum \mathbf{k} and energy ω . The width in energy of the main quasi-particle peak in $A(\mathbf{k}, \omega)$ gives the quasi-particle lifetime at momentum \mathbf{k} . Due to electron correlations additional structures or satellites may be contained in $A(\mathbf{k}, \omega)$. All this can be measured by EMS.

3. EMS of Atoms and Molecules

Advances in EMS of atoms and molecules will occur mainly through significant improvements in the energy resolution and in operating the spectrometers at energies significantly above the 1–1.5 keV currently in use. The essential step in both of these advances is increased sensitivity, which will come with the introduction of multivariable detection techniques (Storer *et al.* 1994). EMS requires high momentum transfer ($K = |\mathbf{k}_0 - \mathbf{k}_f|$) ionising collisions, and since the (e, 2e) cross section goes as K^{-4} , the cross section is very small in the EMS regime. The relatively poor energy resolution of most EMS measurements in the

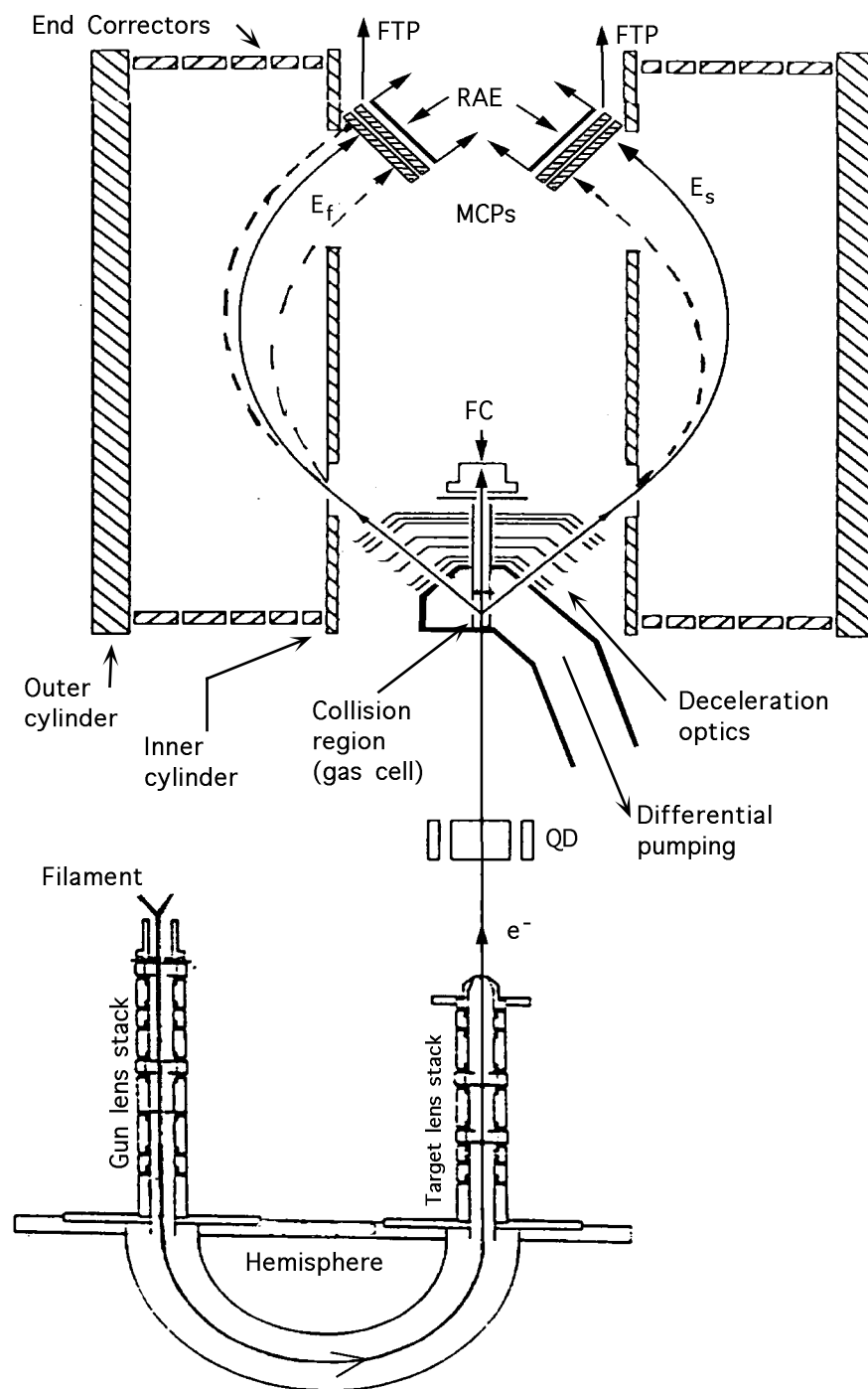


Fig. 1. Schematic of a possible multivariable EMS coincidence spectrometer for gaseous targets based on the cylindrical mirror deflector. It incorporates a high resolution electron source, diametrically opposed micro-channel plate two-dimensional detectors (RAE), fast timing pulses (FTP), and differential pumping.

past is a direct result of this. In order to increase the low coincidence count rate with gaseous atomic and molecular targets the incident current is usually maximised. The resulting space-charge effects broaden both the energy and momentum resolution of the incident beam. The experimental energy resolution of an EMS spectrometer is determined by the addition of each component in quadrature, i.e.

$$\Delta E_{\omega} = (\Delta E_0^2 + \Delta E_f^2 + \Delta E_s^2)^{\frac{1}{2}}. \quad (6)$$

It is usually dominated by ΔE_0 . Brunger and co-workers (Hewitt *et al.* 1998) and Canney *et al.* (1997a) have both used monochromators with thermionic electron emission sources to give much improved energy resolution. Better energy resolution can also be obtained with laser based photo-electron sources. Dorn *et al.* (1997, 1998) have used a negative electron affinity GaAs based spin polarised electron source for (e,2e) studies with significant improvement in the energy resolution. For energy distributions of equivalent widths, these photoemission sources give currents which can be at least ten times greater than those obtained by coupling thermionic emitters with monochromators.

With gaseous samples, target densities can be increased by the use of supersonic nozzles or differential pumping. The EMS spectrometer of Brunger's group (Hewitt *et al.* 1998) already incorporates a differential pumping stage. For condensible vapours this is rather easier, as suitable cold fingers can provide the necessary differential pumping (e.g. Dorn *et al.* 1998).

EMS spectrometers for gaseous targets will need multivariable detectors to gather data simultaneously over a range of energies and momenta. Several possibilities are under consideration (Brunger 1997; Zheng 1997; Chen *et al.* 1997). The double toroidal spectrometer of Miron *et al.* (1997) could be adapted for (e,2e) coincidence spectroscopy in non-coplanar symmetric geometry. Similarly the momentum dispersive spectrometer of Todd *et al.* (1994) can be extended to be also dispersive in energy and include differential pumping as shown schematically in Fig. 1. The monochromated incident electron beam passes along the azimuthal symmetry axis of a cylindrical mirror analyser. The collision region is differentially pumped to increase target gas density. The cylindrical mirror analyser collects electrons over a range of emitted energies, as well as over diametrically opposing segments of azimuthal angular ranges. The two-dimensional microchannel plate position-sensitive detectors (MCPs) are segmented in order to reduce pulse pile-up and to optimise signal to accidental background coincidences. Due to the limited momentum range of valence electrons, $\lesssim 2$ a.u., opposing detector segments each need to cover only a small angular range determined by the kinematics. The segmentation only needs to be done at the anode plate, which can consist of a series of wedge-strip or resistive anode encoders (RAE). Pairs of diametrically opposed detectors can in principle be stacked right around the whole 2π azimuthal range of the spectrometer.

The timing information taken from the back of the MCPs (FTPs) and the charge pulses from the position sensitive detectors (RAEs) can be treated online by a dedicated multiparameter acquisition system and computer, as in Storer *et al.* (1994), to determine the momenta and energies of all the electrons detected within a certain arrival time difference (say 100 ns). From this information

transit time corrections are made to improve the coincidence timing resolution. True coincidence $N_t(\omega, \mathbf{k})$ and accidental background $N_b(\omega, \mathbf{k})$ histograms are then constructed in the usual way, where

$$N_t(\omega, \mathbf{k}) = N_c(\omega, \mathbf{k}) - \frac{\Delta t_c}{\Delta t_b} N_b(\omega, \mathbf{k}). \quad (7)$$

Here N_c are events in the (corrected) timing spectrum within the coincidence window Δt_c , the accidental background window being Δt_b .

The spectrometer response function is given by the accidental background histogram. If the response functions of the two-dimensional position sensitive detectors are $\mathcal{F}(E_f, \mathbf{k}_f)$ and $\mathcal{F}(E_s, \mathbf{k}_s)$ and of the incident beam $\mathcal{F}(E_0, \mathbf{k}_0)$, then

$$N_b(\omega, \mathbf{k}) \propto \mathcal{F}(E_0, \mathbf{k}_0) \star \mathcal{F}(E_f, \mathbf{k}_s) \star \mathcal{F}(E_s, \mathbf{k}_s), \quad (8)$$

where energy and momentum conservation are understood and ' \star ' denotes convolution of the functions with respect to energy and momentum. The form of $\mathcal{F}(E_0, \mathbf{k}_0)$ is open to some choice. For a monoenergetic parallel beam it is just a delta function in energy and momentum. If the electron gun potential is scanned with uniform electron exposure over the range stepped through, then $\mathcal{F}(E_0, \mathbf{k}_0)$ is uniform in energy.

The differential cross section is proportional to the measured true coincidence intensity corrected for the spectrometer response function, i.e.

$$\sigma_{(e, 2e)}(\omega, \mathbf{k}) \propto \frac{N_t(\omega, \mathbf{k})}{N_b(\omega, \mathbf{k})}. \quad (9)$$

The statistical uncertainty in the background spectrum can be significantly reduced by scanning the incident energy over a wide range (Caprari 1994), and for spherically symmetric samples by using the fact that the cross section is independent of the direction of \mathbf{k} .

As systems with more electrons, particularly core electrons, are studied, it will be necessary to increase the energy of the electrons well above the 1–1.5 keV now in general use. This is to ensure that distortion effects are negligible and that the continuum electrons can be adequately described by plane waves. I believe that it will become standard to operate around 5 keV total energy. At this energy, high energy resolution is still easy to achieve. Higher energy operation will bring with it demands for better angular resolution. The Flinders high energy condensed matter spectrometer demonstrates that this can be achieved.

Another development will be the increased use of spin-polarised electron beams and oriented or aligned targets. The dichroism that has recently been observed for (e, 2e) collisions with excited and oriented sodium atoms (Dorn *et al.* 1998) opens up new physics. This work shows that it is possible to measure the direction of rotation of the electron around the nucleus. What will we observe in optically active molecules? We can partially align some of these as well as other molecules by use of supersonic beams, lasers, and the interaction of electric fields with dipolar molecules. Developments in target technology will also include the production of radicals (after all atomic hydrogen has already been used as

a target, Lohmann and Weigold 1981), metastable, and short lived states. The trend to larger and biologically interesting molecules will accelerate.

The future of EMS applied to atoms and molecules looks very bright indeed. This will come about by straightforward developments in spectrometers, incident beams, and target preparation.

4. EMS of Condensed Matter

Several of the developments discussed for the case of gaseous targets are also relevant for solid state targets. However, the technical problems are sufficiently different to deserve special consideration.

EMS applied to solids shows great promise. Vos (1998, present issue p. 609) gives an outline of the remarkable progress that has occurred in the past few years in the transmission measurements with the Flinders high energy asymmetric spectrometer. The advantages of EMS applied to condensed matter include:

(1) Direct measurement of $A(\mathbf{k}, \omega)$ with no integration over energy or momentum. (2) Momentum direction cuts can be selected. (3) Surface sensitivity due to the small mean free path of the slow electron at lower energies. This can be adjusted to some extent by tilting the sample to change the angle of the slow electron relative to the surface. (4) It yields $A(\mathbf{k}, \omega)$ for amorphous and polycrystalline material, as well as single crystals. (5) The instrumentation and running costs are relatively low.

In the normal arrangement of the Flinders spectrometer $A(\mathbf{k}, \omega)$ is measured along an axis in momentum space which is normal to the incident beam direction and passes through the Γ point ($\mathbf{k} = 0$). By adjusting the polar angle of the slow electron analyser, different cuts can be taken through momentum space. This is demonstrated vividly in the case of a single crystal graphite target (Vos *et al.* 1997), where the cut through the Γ point is chosen to lie in the nodal plane of the pi band. There is therefore no evidence of pi electrons in this measurement. As soon as the cut is shifted outside this plane the pi band becomes visible with finite density. By rotation of the crystal target they are able to measure the differences in dispersion of the sigma bands along the $\Gamma - M$ and $\Gamma - K$ directions. EMS is currently the only spectroscopy which can show in which Brillouin zone a particular band has non-zero momentum densities.

The surface sensitivity of EMS has been demonstrated in a number of measurements, the most recent of which is the study of the oxidation of aluminium (Canney *et al.* 1997b). The measured spectral-momentum densities show how those of aluminium oxide begin to dominate over the free-electron band of metallic aluminium as the exposure to oxygen is increased.

There are, however, some technical difficulties in the case of solids. These result from the high electron densities of the target. Because of the relatively short mean free path of low keV electrons in condensed matter, multiple scattering effects are prominent. Elastic scattering of the incident or emitted electrons, respectively before or after the (e, 2e) event, will result in a derived momentum \mathbf{k} from (equation 1) which is incorrect due to the unknown momentum transfer in the elastic scattering process. The binding energy of the electron will be preserved, but due to the vector nature of the momentum conservation equation, there will be a background of true coincidences smeared out over the measured

momentum range. Inelastic scattering of the continuum electrons, mainly due to plasmon excitation in the target sample, causes a shift in energy of $A(\mathbf{k}, \omega)$. This can be corrected for to some extent by measuring the energy loss spectra for electron beams of energies E_0, E_f and E_s using the spectrometer analysers and then semi-empirically deconvoluting the data (Dennison and Ritter 1996). However, deconvolution of the data is prone to errors, and satellite structures due to electron correlation effects in the target material can be concealed by larger inelastic contributions. Vos and Bottema (1996) discuss the degradation in contrast in the measured spectral-momentum densities which results from these multiple scattering effects. They introduce a Monte-Carlo simulation technique, which from first principles shows how a known (calculated) $A(\mathbf{k}, \omega)$ will be modified by multiple scattering. Their Monte-Carlo simulations agree very well with the measured data.

In order to measure $A(\mathbf{k}, \omega)$ for all \mathbf{k} one is required to use the transmission mode of measurement. Due to the prominence of multiple scattering, this means that the targets have to be very thin self-supporting membranes ($\lesssim 20$ nm). The preparation of such thin self-supporting targets is difficult, and different materials bring with them different problems. However, significant advances have been made in target sample preparation and characterisation techniques (Fang *et al.* 1997), and more advances can be expected. Thus the range of target material does not appear to be a limiting factor.

Multiple scattering effects and difficulties in target preparation can be minimised by the simple expedient of going to higher energies and symmetric final state kinematics. The difficulty in going to higher energies and symmetric kinematics is in maintaining good energy resolution (equation 6). This requires ripple and drift free power supplies for setting the voltages defining E_0, E_f and E_s . An additional difficulty is that the (e, 2e) cross section goes down as K^{-4} . The drop in cross section is, however, largely compensated for by the increase in the effective thickness of the target sampled due to the increase in outgoing energies.

Such a high energy spectrometer, with $E_0 = 60$ keV and $E_f = E_s = 30$ keV, is under construction at the ANU. It is shown schematically in Fig. 2. The electron gun is designed to float at -30 kV and the whole collision region is at the uniform potential of $+30$ kV. This aspect of the design is similar to that of the single channel instrument of Ritter *et al.* (1984).

It has the advantage of having the hemispherical energy analysers, the channel plates and the two-dimensional position-sensitive anodes near the ground potential. The target chamber, with a hemispherical exit surface, sits inside the vacuum chamber with a hemispherical end and floats at the high positive potential. The hemispherical analysers behind the conical slit decelerating lenses accept electrons with polar angles of 44° over a range of azimuthal angles and energies. The polar angle of 44° allows for relativistic effects and finite binding energy, ensuring that the momentum range probed passes through the $\mathbf{k} = 0$ point. The actual polar angles θ_f and θ_s selected can be varied about 44° by pairs of electrostatic deflectors placed at the exit of the target chamber. In this way different cuts can be taken through momentum space. The spectrometer design allows for the addition of two more diametrically opposed analysers perpendicular to the ones shown.

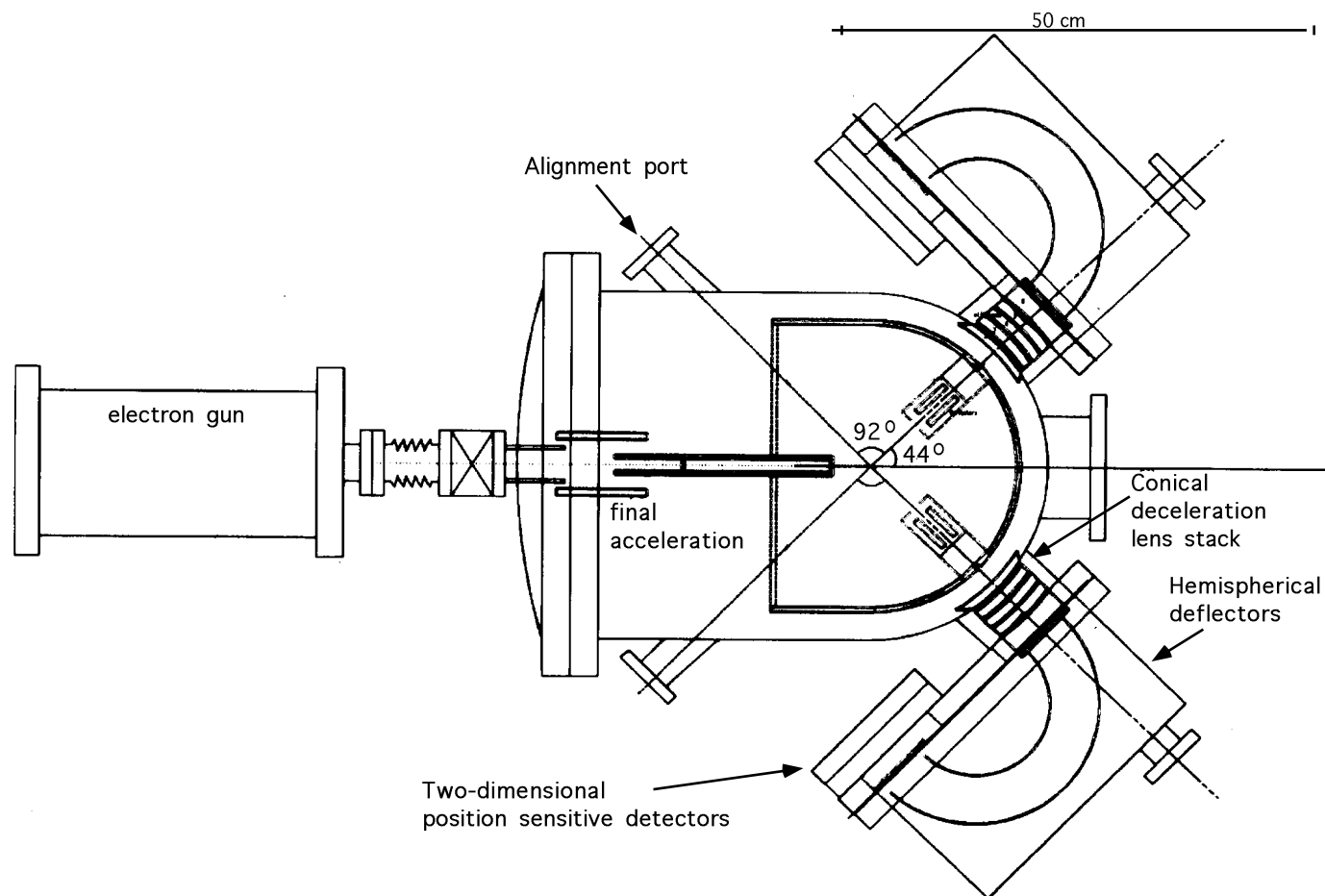


Fig. 2. The new high energy (e,2e) spectrometer at the ANU. The monochromated electron gun and the target chamber float at -30 kV and $+30$ kV respectively.

Specially designed power supplies, with long-term relative stabilities of the order of 1×10^{-5} , will give a long-term energy resolution of the order of 0.5 eV. Higher energy resolution for looking at special effects, say near the Fermi level, should be possible at lower total energies.

At larger binding energies the energy resolution is not a limiting factor because quasi-particle lifetime broadening of the energy bands dominates. This lifetime broadening results from electron–electron correlation effects. These effects are also predicted to give rise to additional structure with significant intensity at higher binding energies (Lundqvist 1968; Aryasetiawan *et al.* 1996). EMS, especially with a reduction in multiple scattering effects, will permit direct measurement of these correlation effects and hence provide stringent tests for many-body theories of condensed matter.

Higher energy operation not only significantly reduces multiple scattering effects with a resulting improvement in the clarity of the data, but it will also make target preparation easier. It will be possible to use thicker target samples without deleterious effects on the signal to noise ratio. Of course, if the target samples are too thick, multiple scattering will again become a problem.

For the ANU spectrometer we are also planning to introduce a spin-polarised electron source. As well as the advantage of good energy resolution, the use of spin polarised incident electrons should reveal much new information, particularly on magnetic phenomena and relativistic effects. EMS on superconductors promises to be a most exciting area of investigation. If work on molecules is any guide, the long-range correlations in superconductors could show up as significant extra density at low momenta. It would be fascinating to directly measure the square of the electronic wavefunction of matter in the superconducting state. Other strongly correlated systems, metals, alloys, surfaces and semiconductor crystals are all waiting to be exploited.

5. Summary

EMS of atoms and molecules, although in some respects already a mature field, is on the threshold of major developments. Multivariable detector techniques, improved energy resolution, spin-polarised incident electrons, polarised and oriented targets, and higher energy operation, will all lead to major new applications. The application of EMS to larger molecules and biologically interesting molecules promises to be particularly rewarding (Neville *et al.* 1996). EMS can provide orbital-specific information regarding the quality of a particular wavefunction, particularly in the chemically interesting bonding region. It thus provides a stringent basic test for alternative quantum chemistry computational techniques. The information obtained from EMS should be valuable for improving the input to computer-aided molecular design studies of structure and reactivity of very large molecules such as proteins. With optical pumping techniques or supersonic nozzles and field-guided molecular beams it should become possible to determine three-dimensional orbital momentum densities of spatially aligned molecules.

EMS of solids is in its infancy. It promises to be a most exciting area of research. High energy machines will reveal details of correlation effects in their measurement of the quasi-particle spectral momentum densities $A(\mathbf{k}, \omega)$ in strongly correlated systems. These can be single crystals, polycrystalline, or amorphous. Superconducting and magnetic effects can be directly measured. Surfaces and

surface adsorbates and adsorbates can also be selectively chosen for investigation, and the alignment and orientation of surface molecules directly measured.

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