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Electron–Auger Electron Coincidence Experiments: Current Status and Future Prospects*

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Abstract

Approximately ten years ago the first experiments were performed in which the Auger electrons produced after inner-shell ionisation of atoms by electron impact were detected in coincidence with the scattered electrons. Only a limited number of such experiments have been performed since that time, mainly due to the very low count rates characteristic of these measurements. Recent developments in the field are discussed and the future prospects for such measurements are considered.

1. Introduction

Electron impact ionisation of an atom by the knock-out of an inner-shell electron leaves the ion in an excited state. If the target is a low Z atom, then the ion will subsequently decay by emitting an Auger electron. The process may be represented as follows:

$$
\begin{aligned}
e_{\text{incident}} + \mathrm{A} \rightarrow \mathrm{A}^{+*} &+ e_{\text{ejected}} + e_{\text{scattered}} \\
&\rightarrow \mathrm{A}^{++} + e_{\text{Auger}}\,. \qquad (1)
\end{aligned}
$$

Hence, in the outgoing channel there are four charged particles, the residual ion and the three electrons. Many experiments have been performed in which only the Auger electron is detected after the collision, and a wealth of information can be obtained from such measurements. However, the measured cross sections are essentially an average over all possible partitions of the energy and momentum between the two undetected electrons. To fully specify the particular collision kinematics would require the detection, in coincidence, of all three outgoing electrons. Although triple-coincidence experiments have been performed for the case of outer-shell ionisation (Lahmam-Bennani *et al.* 1989), the very low cross sections for inner-shell ionisation mean that such experiments, for the process given in equation (1), are at the limits of what is achievable with current technology. The first steps towards such a 'complete' experiment were taken by Sewell and Crowe (1982, 1984*a*, *b*) and Sandner and Völkel (1984) in pioneering experiments

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in which they detected the Auger electron in coincidence with a scattered electron of specified energy and momentum [often referred to as $(e, e'e_{\mathrm{Auger}})$ experiments]. The energy of the ejected electron is thereby fixed since

$$E_0 = E_{\mathrm{sc}} + E_{\mathrm{ej}} + \mathrm{IP}\,, \tag{2}$$

where E_0 is the incident energy, E_{sc} and E_{ej} are the energies of the scattered and ejected electrons respectively and IP is the ionisation potential of the electron in the inner-shell orbital. A limited number of groups have performed $(e, e'e_{\mathrm{Auger}})$ experiments since that time, and in this paper I give a brief overview of these measurements. Although a wide range of experiments involving Auger electrons has been performed, I restrict the discussion in this paper specifically to the electron–Auger electron coincidence experiments.

Most of the experiments which have been performed to date can be conveniently divided into two categories: (1) lineshape measurements (a measurement of the Auger yield as a function of energy) and (2) angular correlation measurements (a measurement of the Auger yield as a function of angle). Before discussing these in detail it is, however, appropriate to mention the experiment of Doering *et al.* (1990) in which they measured coincidences between Auger electrons and ejected electrons of the same energy, as a function of incident energy. These measurements do not really fall into either of the former categories, but they provided some thought-provoking evidence of strong electron correlation, particularly near threshold.

Fig. 1 illustrates a typical experimental configuration used for electron–Auger electron coincidence measurements.

2. Lineshape Measurements

The first coincidence lineshape measurements were reported by Sewell and Crowe (1984*a*). They measured the yield of Ar $L_2M_{23}M_{23}(^3P_{012})$ Auger electrons as a function of energy, in coincidence with scattered electrons of a given energy. Both the scattered and Auger electrons were detected at a fixed angle of emission. The experiment was performed under conditions where the (undetected) ejected electron is constrained to have a rather low energy ($\sim$5 eV) compared with the energy of the Auger electron (207·2 eV in this case). Sewell and Crowe found that the lineshape measured under these conditions was shifted to a higher energy and broadened on the high energy side, when compared with the Auger lineshape obtained by detecting only the Auger electron (i.e. not in coincidence with the scattered electron). The shift was attributed to the Coulomb interaction between the slow ejected electron (which is still in the vicinity of the ion when the Auger decay takes place) and the Auger electron emitted subsequently. Similar effects had been observed in Auger decay after both electron impact ionisation and photoionisation, and the phenomenon was dubbed post collision interaction (PCI) (Schmidt 1992). However, in these previous experiments, only the Auger electrons were detected. In the case of electron impact ionisation, if the Auger electrons are not measured in coincidence with the scattered electrons, then the measured lineshape is an average over all possible energy-sharings of the ejected and scattered electrons. Hence, unless the experiments are performed near threshold, there is only a small contribution to the lineshape from those events where the ejected electron has a very low energy. Nevertheless, careful measurements (Völkel *et al.* 1988), as well as calculations (Sandner and Völkel 1989), have shown that there is always a residual energy shift of the Auger line (albeit small—about 14 meV) if the Auger electrons are not measured in coincidence. Only if a coincidence measurement is performed can the ejected electron energy be constrained to be much greater than the Auger electron energy, ensuring that no PCI can take place. Thus the coincidence experiments, because they enable one to select specific kinematic conditions, can have the effect of accentuating or removing PCI effects in the observed lineshapes.

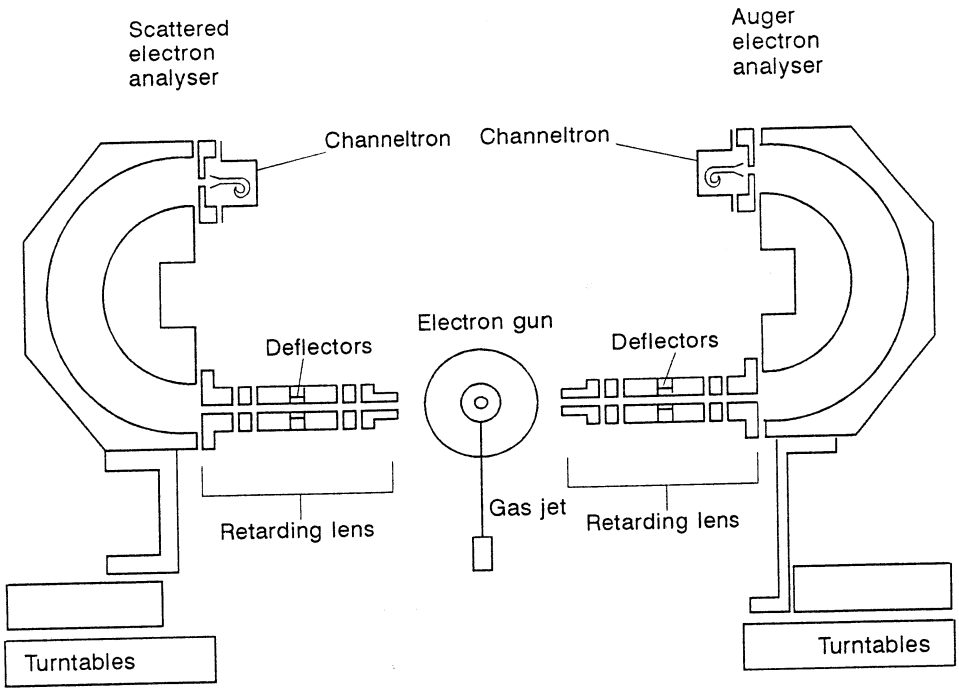

Fig. 1. Typical experimental configuration for coplanar electron–Auger electron coincidence experiments.

Further measurements of the effect of PCI on the Auger lineshape have been reported by Sandner and Völkel (1984), Stefani *et al.* (1986), Lohmann (1991) and Weigold (1992). Fig. 2 illustrates the shift to higher energy and broadening of the lineshape which is characteristic of the post collision interaction. In addition to energy and angular momentum exchange due to PCI, the Auger lineshape may also be affected by the presence of a second channel to the final doubly-ionised state (for example a direct double ionisation). If such a channel exists, then there is a possibility of quantum mechanical interference between the two amplitudes. Evidence for such interference phenomena has been observed by Sandner and Völkel (1984), Lohmann (1991) (see Fig. 3) and Avaldi *et al.* (1993).

A number of theoretical models of PCI have been developed, including those of van der Straten *et al.* (1988), Kuchiev and Sheinerman (1989, 1994) and Armen *et al.* (1987), and the lineshapes predicted by these models generally show good agreement with the experimental data. The confirmation of the presence of PCI in the final channel means that the Auger process may no longer be treated as a two-step process, in which the initial ionisation and the subsequent decay of the ion are treated as independent events.

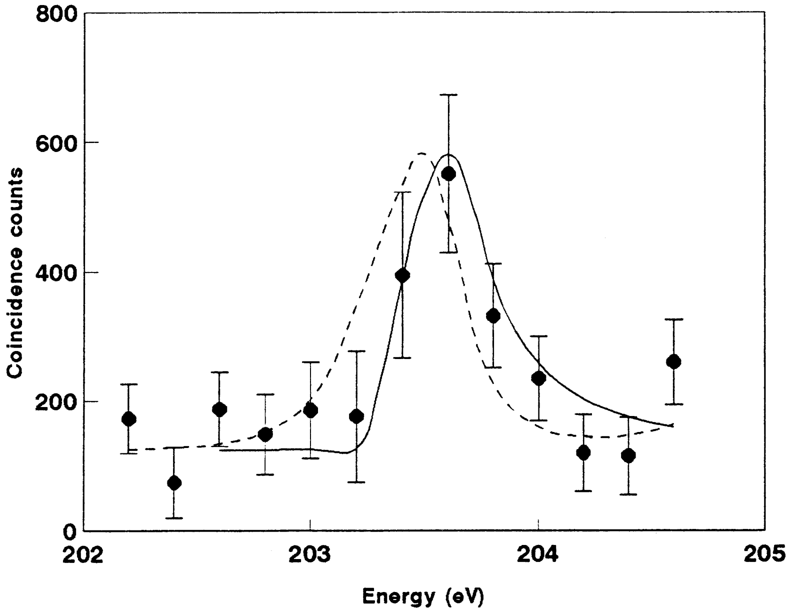

Fig. 2. Coincidence spectrum of the argon $L_3M_{23}M_{23}(^1D_2)$ Auger line at an excess energy of 5 eV; the broken curve is the non-coincident spectrum (from Lohmann 1991). The solid curve is the Kuchiev and Sheinerman lineshape function (see equation 3) evaluated for an excess energy of 5 eV and convolved with the experimental energy resolution.

The theoretical formulation of PCI due to Kuchiev and Sheinerman yields a lineshape function of the form

$$P(\epsilon_A) = \frac{\Gamma/2\pi}{(E_A^0 - \epsilon_A)^2 + \Gamma^2/4}\,\kappa(\epsilon_A, \xi)\,, \tag{3}$$

with

$$\kappa(\epsilon_A, \xi) = \frac{\pi\xi}{\sinh(\pi\xi)}\exp\left(2\xi\tan^{-1}\frac{2(E_A^0 - \epsilon_A)}{\Gamma}\right), \tag{4}$$

$$\xi = \frac{1}{k_{sc}} + \frac{1}{|\boldsymbol{k}_{sc} - \boldsymbol{k}_A|} - \frac{1}{k_{ej}} + \frac{1}{|\boldsymbol{k}_{ej} - \boldsymbol{k}_A|}\,. \tag{5}$$

Here $P(\epsilon_A)$ is the probability of detecting an Auger electron with energy ϵ_A, where E_A^0 is the 'nominal' Auger energy and Γ is the initial state width. The parameter ξ includes terms which depend not only on the energies of the three outgoing electrons, but also on their relative directions. In the case where the scattered electron has a much higher energy than the ejected or Auger electron,

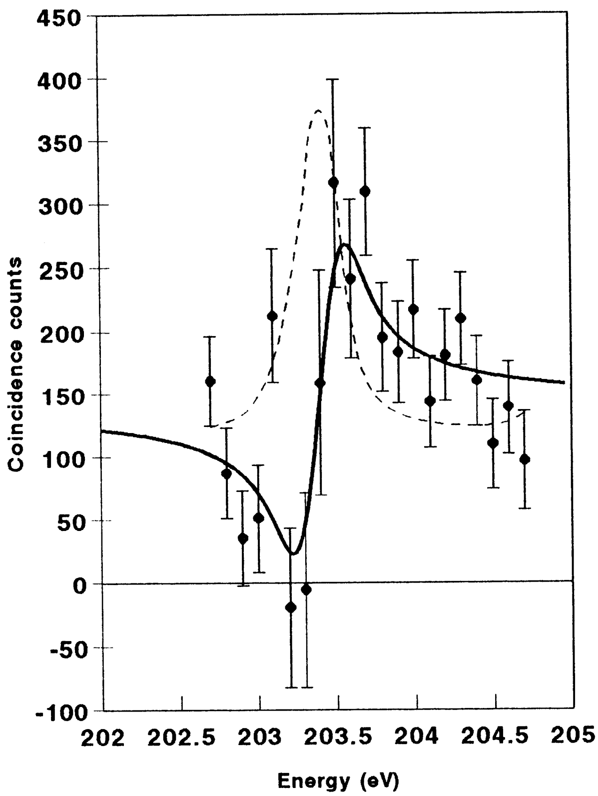

Fig. 3. Coincidence spectrum of the argon $L_3M_{23}M_{23}(^1D_2)$ Auger line at an excess energy of 2 eV. The broken curve is the non-coincident spectrum and the full curve is a fit of a Beutler–Fano profile to the coincidence data (Lohmann 1991).

its influence in the outgoing channel may be neglected; however, the fourth term in (5) still contains a dependence on the angle between the ejected and Auger electrons. Equation (3) represents a normalised lineshape function, and hence the integrated intensity of the Auger line remains constant as the parameters change. Kuchiev and Sheinerman (1994) have recently performed calculations of the effect of PCI on the intensity of the Auger lines as a function of the angle between the photoelectron and the Auger electron after photoionisation. The calculations predict a strong variation in the line intensity at small angles between the two outgoing electrons.

A number of attempts have been made to observe the predicted angular dependence of the PCI. Schnetz and Sandner (1992) measured Ar $L_3M_{23}M_{23}(^1S_0)$ Auger electrons in coincidence with scattered electrons under conditions where the ejected electrons were expected to be concentrated in a direction perpendicular to the direction in which the Auger electrons were detected. They observed a *smaller* PCI effect than for the case where the ejected electron distribution was expected to overlap substantially the direction of Auger emission. This result is in qualitative agreement with the formulation of Kuchiev and Sheinerman (1989), since the effect of the asymmetry parameter becomes larger as the angle between the ejected electron and the Auger electron decreases. The observed effect was very small, but provided some evidence for an angular dependence of the PCI.

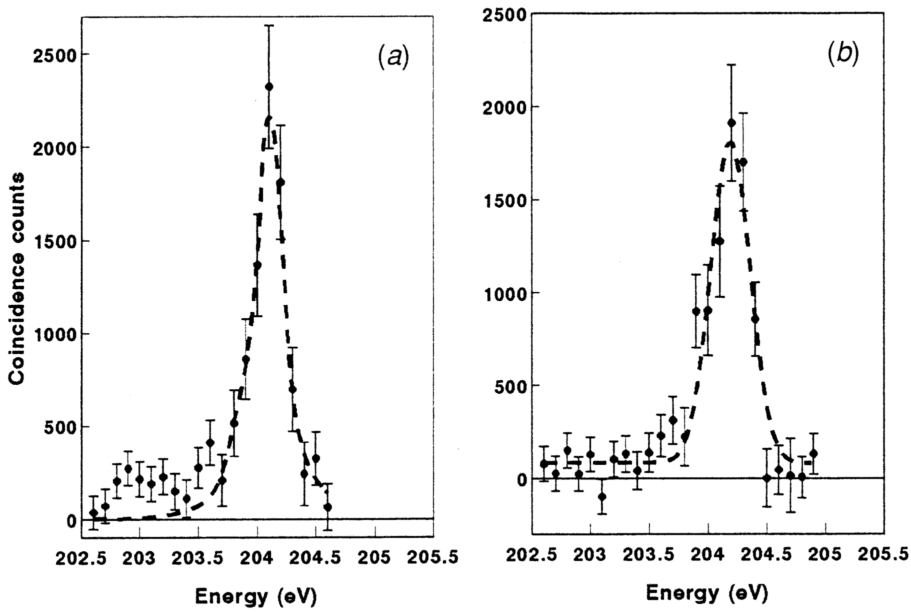

Fig. 4. (a) Coincidence measurement of the $L_3M_{23}M_{23}(^1D_2)$ line in argon at an excess energy of 207 eV. The Auger electron analyser was positioned at an angle corresponding to the maximum in the theoretically predicted ejected electron distribution. The broken curve is the non-coincident spectrum. (b) As for (a) but with the Auger electron analyser positioned at an angle corresponding to the minimum in the theoretically predicted ejected electron distribution (Lohmann *et al.* 1992).

The experiments of Schnetz and Sandner were performed at an excess energy (i.e. the kinetic energy of the ejected electron) much lower than the Auger energy. Under these conditions a simple physical picture of the process involves the Auger electron catching up and overtaking the ejected electron, with the strongest interaction occurring when the two are at the same distance from the nucleus. Inspection of the calculated behaviour of the Auger lineshape as a function of ejected electron energy (see for example Armen 1988) reveals that a very strong shift and distortion of the line to *lower* energy is expected if the ejected and Auger electrons have almost the same energy and are emitted in approximately the same direction. Lohmann *et al.* (1992) thus tried a different approach to investigating the angular dependence of the PCI. They measured the Ar $L_3M_{23}M_{23}(^1D_2)$ Auger electrons in coincidence with the scattered electrons, but under conditions where the ejected electron had almost the same energy as the Auger electron (203·4 eV). Using theoretical calculations of the ejected electron distribution, they measured the lineshape for two different Auger emission directions, one corresponding to a (predicted) maximum in the ejected electron distribution, the other to a minimum. The lineshape measured at a position corresponding to the maximum in the distribution was found to have an unshifted component as well as a small structure on the low energy side (Fig. 4*a*). This structure was absent in the lineshape measured at the other angle (Fig. 4*b*). The results can be interpreted as an indication of an angular dependence in the PCI, with those ejected electrons having trajectories close to that of the Auger electron producing a large PCI shift, while those travelling in directions far from the Auger direction produce almost none. The resultant lineshape is then a sum of these contributions.

The experiments of both Schnetz and Sandner (1992) and Lohmann *et al.* (1992) suffer from the fact that the ejected electron direction is not exactly specified, and hence the resulting lineshapes are in fact an average over the actual ejected electron distribution. This tends to obscure any angle-dependent effects. However, new experiments have very recently been performed (Avaldi *et al.* 1995) in which the Xe $N_5O_{23}O_{23}(^1S_0)$ Auger electrons have been detected in coincidence with *ejected* electrons of nearly the same energy. Avaldi *et al.* were able to measure the Auger lineshape under these conditions with an angle of only 25° between the two electrons. The measured lineshape exhibits a clear shift to lower energies, while a lineshape measured with an angle of 170° between the ejected and Auger electrons exhibits no shift, thus providing clear evidence for an angular dependence of the PCI.

3. Angular Correlation Measurements

The interest in performing angular correlation measurements of Auger electron emission was prompted by the suggestion of Berezhko *et al.* (1978) that such experiments could be used to investigate the alignment of the intermediate ion state i.e. the population of the magnetic sublevels. The alignment of the ion is manifested as an anisotropy in the angular distribution of the decay products. In the case where the two-step approximation holds the angular distribution of the Auger electrons $W(\theta,\phi)$ can be written (Berezhko *et al.* 1978)

$$W(\theta,\,\phi) = \frac{W_0}{4\pi}\left[1 + \sum_k \alpha_k \sum_\kappa A_{k\kappa}\left(\frac{2\pi}{2k+1}\right)^{\frac{1}{2}} Y_{k\kappa}(\theta,\,\phi)\right], \tag{6}$$

where the α_k are proportional to the reduced matrix elements of the particular Auger transition and the $A_{k\kappa}$ are the statistical tensors which describe the ionisation process.

However, the presence of a post collison interaction in the outgoing channel means that calculations based on a simple two-step model plus the first Born approximation, such as that discussed by Berezhko *et al.* are unlikely to be realistic. Subsequent work by Berezhko and Kabachnik (1982) has shown that a distorted wave Born approximation (DWBA) calculation gives substantially different results, even without the inclusion of PCI.

The first Auger angular correlation measurements were performed by Sewell and Crowe (1982, 1984*b*) and Sandner and Völkel (1984) on the $L_3M_{23}M_{23}(^1S_0)$ Auger line in argon. The results from the two contemporary measurements were not in agreement, although both groups concluded that the simple two-step Born approximation calculation was inadequate. Stefani *et al.* (1986) also measured

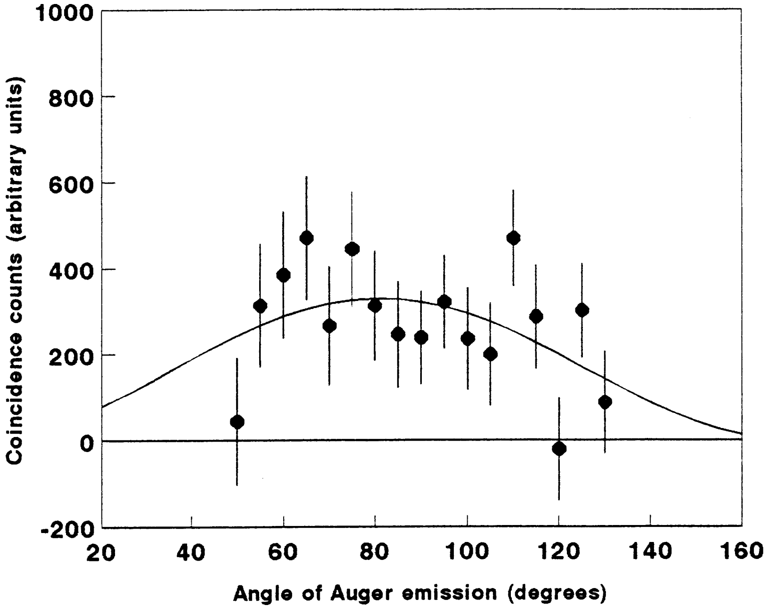

Fig. 5. Preliminary angular correlation measurement of the argon $L_3M_{23}M_{23}(^1D_2)$ Auger electrons, at an excess energy of 5 eV. The curve is a fit of equation (7) (see text).

the angular correlation of the Ar $L_3M_{23}M_{23}(^1S_0)$ Auger electrons, but at much higher energies (8 keV compared with 1 keV). Again, their results confirm that the models based on a two-step process plus a first Born approximation are not appropriate, particularly if PCI and interference effects are present. The question of what information can be obtained from angular correlation measurements in the regime of low excess energies is still an open one. Clearly new theoretical calculations are needed, however the provision of additional experimental data is probably a prerequisite. Only a handful of other angular correlation measurements have been performed since then, and none of the data has been published. We have performed a couple of measurements of the angular correlation of the Ar $L_3M_{23}M_{23}(^1D_2)$ line at a scattering angle of 20°, ejected electron energy of 5 eV and scattered electron energy of 750 eV. The two sets of measurements have been added to produce the angular correlation presented in Fig. 5. One set of measurements (measured over the angular region from 55° to 105°) corresponds to data obtained using a position sensitive detector to detect the Auger electrons (Johnson *et al.* 1994). This enabled us to measure the whole Auger lineshape at each angle, as well as providing a measure of the background. The second set of measurements (measured over the angular range from 50° to 130°) was obtained by measuring the coincidence signal alternately at the peak of the Auger line and 1 eV below the peak (to get a measure of the background at each angle). The latter set of data was obtained using a channeltron to detect the Auger electrons. The two data sets are consistent over their common angular range, but the data must still be considered preliminary.

The coplanar angular correlation may be described (Berezhko and Kabachnik 1982) by a function of the form

$$W(\theta) \propto 1 + \beta_{||} \cos[2(\theta - \psi)]\,, \qquad (7)$$

where $\beta_{||}$ is a measure of the anisotropy of the distribution and ψ indicates the angular position of the minimum in the distribution. If the Born approximation holds then ψ coincides with the momentum transfer direction θ_K (=58° for our measurements).

Although the presence of an angular dependent post-collision interaction may result in a departure of the real angular distribution from that predicted above, equation (7) serves as a useful means of comparing the angular correlation data from different measurements. A fit of equation (7) to our data yields $\beta_{||} = -0\cdot99{\pm}0\cdot56$ and $\psi = 170\cdot9°{\pm}10\cdot6°$. We know of only one other angular correlation measurement of this line, preliminary results of which were reported by Weigold (1992). Although this measurement was performed under somewhat different experimental conditions than ours, and hence the results are not directly comparable, a nonzero anisotropy and a displacement of the minimum in the distribution from the momentum transfer direction was also found.

4. Conclusions

The coincident detection of Auger electrons and scattered (ejected) electrons after inner-shell electron impact ionisation has been shown to yield new and interesting results. There has been substantial progress in theoretical descriptions of the process under study, particularly the effects of post collision interaction on the energy and angular distribution of the Auger electrons. There are a number of promising avenues for future experimental work, including a study of the cross-over from a positive PCI energy shift to a negative PCI energy shift of the Auger line, the influence of angular momentum transfer in the post collision interaction and the intensity variation of the Auger lineshape with relative angle for the case where the ejected and Auger electrons have similar energies.

In the case of Auger electron angular correlation measurements, there is a need for further reliable measurements, particularly a systematic study of the correlations as a function of varying scattering angle and excess energy, and for different Auger lines. There is clearly a need to separate out the contribution to the distribution from any intrinsic anisotropy of the Auger decay, from the collision-induced anisotropy and from PCI effects in the final channel.

Continuing improvements in the technology of coincidence measurements offer the prospect of addressing these problems in the near future.

Acknowledgments

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