

Angular Correlations in Electron-Atom Scattering

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Abstract

The principles of angular correlation coincidence experiments are discussed in relation to electron impact excitation of atoms. Examples are given of the resolution of triplet states above the 14.7 eV dissociation limit of H₂ and of the determination of differential cross sections for the magnetic sublevels of the 2p state of atomic hydrogen.

Introduction

The information obtained from collisions between atomic particles depends on the type of particles (electrons, photons, atoms or ions), on the kinematics of the particles (momentum vector, energy, time of emission from the collision system) and on the internal forces (as in molecules or solids) as well as the external forces (electric or magnetic fields) acting on the collision system. Information is obtained about atomic structure (before, during and after a collision), about total and differential cross sections and about scattering amplitudes and their phases. In molecules, the angular and kinetic energy distributions of dissociation products provide knowledge of the symmetry and internal energy of the dissociating parent states.

Recent advances have been made using coincidence techniques to study atomic lifetimes via sequential cascading photon detection, to specify collision dynamics by correlating scattered particles in multiple decay modes and to deduce scattering amplitudes and relative phases through angular correlation measurements. This paper will only be concerned with electron impact excitation processes and the coincident detection of the scattered electron and decay photon. A more detailed treatment of angular correlation phenomena would include electron impact (e, 2e) ionization processes (McCarthy and Weigold 1976), ion-atom collision processes (e.g. Vassilev *et al.* 1975; Eriksen and Jaecks 1978; Zehnle *et al.* 1978) and beam-foil collision processes (Berry 1977).

Scattering experiments are being refined to produce and detect pure quantum states; for example, an electron with a set of quantum numbers ($nljm$) or a photon beam of intensity I , momentum vector hk and helicity. Progress is illustrated with the following examples. Rubin *et al.* (1969) used a polarized sodium atom beam in a recoil-type experiment to determine the ratio of spin-flip to total differential cross sections for electron-alkali inelastic scattering. Hanne and Kessler (1974) used

polarized electrons to study electron exchange scattering from mercury atoms. Hils and Kleinpoppen (1979) scattered polarized electrons from polarized potassium atoms to observe interference effects in ionizing collisions. Each of these experiments reduces the number of collision parameters over which an average or summation must be made in order to uniquely define the perfect scattering experiment and so enable the most basic quantities of amplitudes and phases to be measured and compared with theory (Burke and Williams 1978).

This short paper is further restricted to just two recent examples of the use of the coincidence technique in electron impact excitation studies, namely the identification of H_2 molecular dissociation modes and coherent excitation of $H(2p)$ magnetic sublevels.

Electron Spectroscopy

A good high resolution electron scattering apparatus permits an incident electron beam energy from 10 to 1000 eV with a resolution of 20 meV and the simultaneous detection of scattered electrons with an energy resolution of 20 meV and of emitted photons of wavelength from 1000 to 50 nm with better than 2 nm bandwidth except at the shorter wavelengths. Optical and energy-loss excitation functions are readily obtained. The capabilities of the techniques are best illustrated by measurements for molecular hydrogen in the region of states with binding energies from 11 to 16 eV, as shown in Fig. 1. Attention is focused on optically forbidden transitions which are frequently strong for a few electron volts above threshold where resonances can dominate. The recent results of Bose and Linder (1980) show, in Fig. 2, a constant residual-energy electron spectrum clearly identifying the dominant $c^3\Pi_u$ state and probably other triplets $d^3\Pi_u$, $e^3\Sigma_u^+$, $f^3\Sigma_g^+$, $g^3\Sigma_g^+$, $k^3\Pi_u$, $m^3\Sigma_u^+$ and $n^3\Pi_u$ near the dissociation and ionization limits. Fig. 3 shows a typical excitation function obtained by electron energy loss measurements for a triplet state, the example being the $c^3\Pi_u$ state for $v = 0, 1$ and 2 . Similarly one can obtain optical excitation functions by photon production, as shown in Fig. 4 for VUV photons within the bandwidth 112–130 nm. Resonances due to the c and a states are seen near 12 eV and the onset of higher predissociating triplet states are seen near 14.7 eV. The resonances are well known and arise when there is sufficient interaction in the incoming channel to excite the target atom and when the slow outgoing electron forms a resonant or virtual state in the excited state of the molecule. This enhancement arises from final state interactions. A similar model was used by Linder (1978) to explain threshold resonances in polar molecules.

The role of the triplet contribution to the 112–130 nm photon signal can be resolved by coincidence measurements of those photons with the near-zero-energy scattered electrons, as shown in Fig. 5. Coincidences are detected for all prominent triplet excitations above the 14.7 eV dissociation limit of $H(2p) + H(1s)$, whereas triplet excitation below 14.7 eV gives only random coincidences. It is apparent that the triplet states above 14.7 eV predissociate giving Lyman- α photons. Such an experiment indicates the role of a coincidence technique in sorting out the decay mechanisms of $e-H_2$ interactions near 15 eV. However, an extension of these measurements is required to elucidate the collision dynamics and excited state description, as is indicated in the next section for atomic hydrogen.

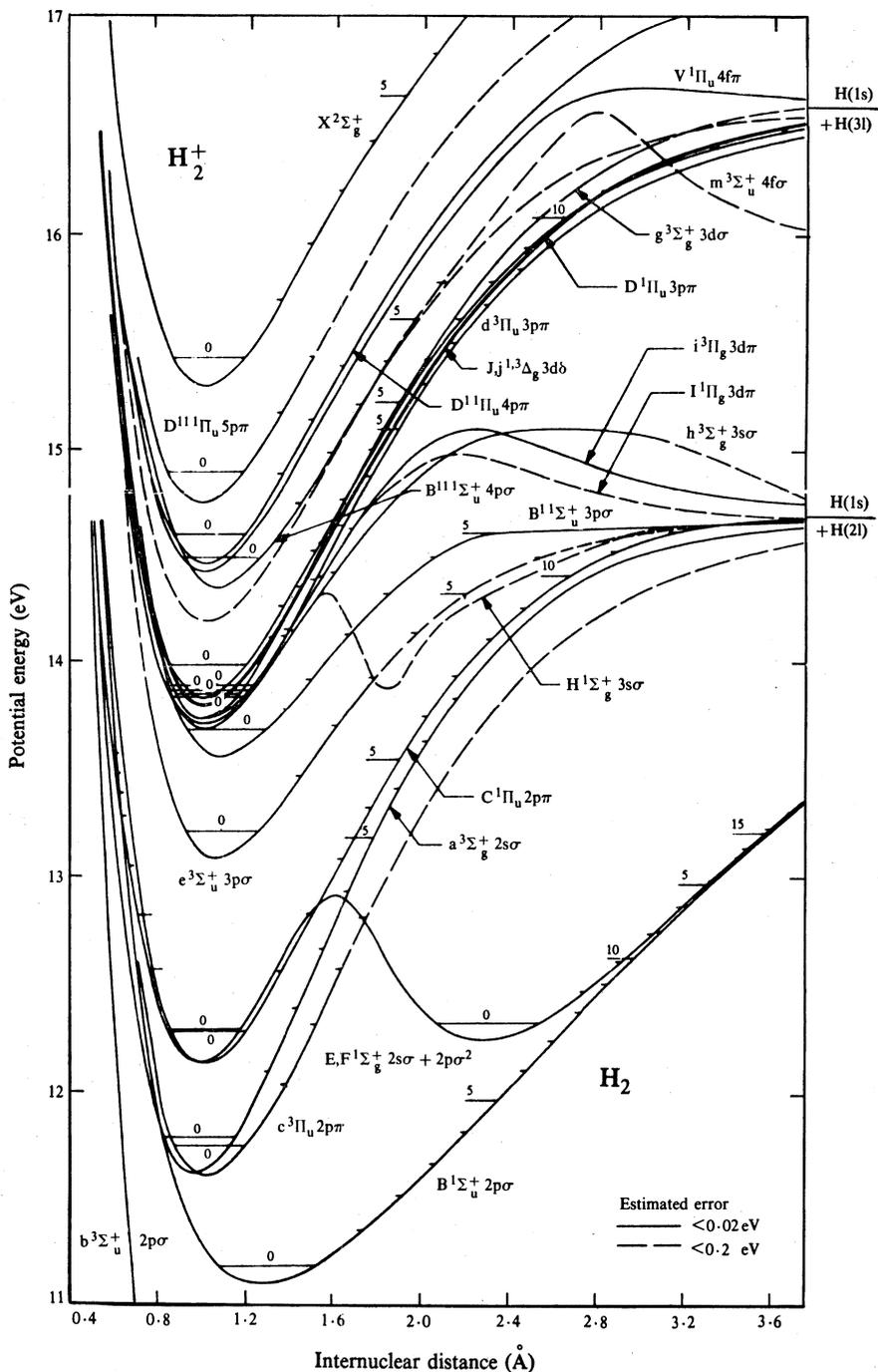


Fig. 1. Potential energy curves for H_2 and H_2^+ (from Sharp 1971).

Excited State Multipoles

The theory of the measurements concerns the relationship between the geometrical properties of the radiation as well as the parameters which characterize the atomic source. Successive developments of this theory have been given by Fano (1957), Macek and Jaecks (1971), Fano and Macek (1973), Gabrielse and Band (1977) and Blum and Kleinpoppen (1979). Only the basic results, mainly following Blum and Kleinpoppen are given here.

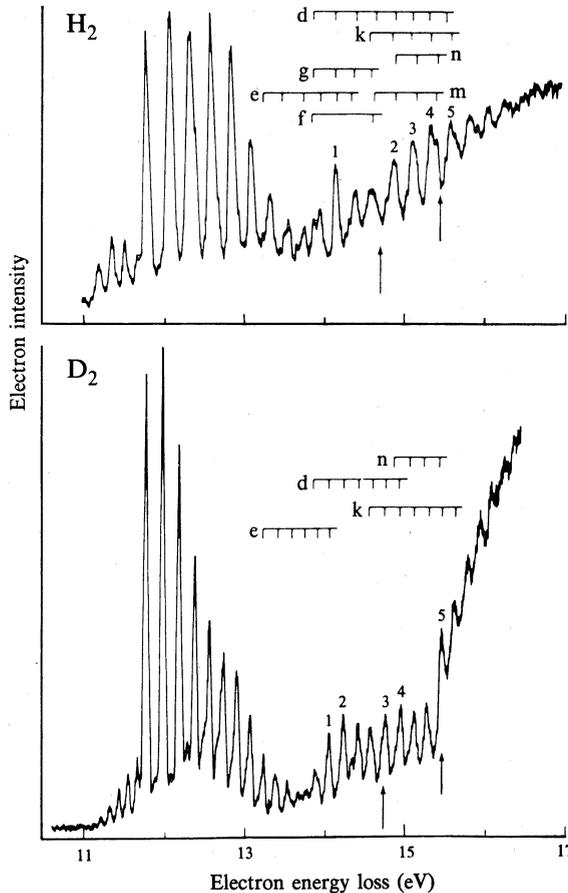


Fig. 2. Constant residual-energy (E_{res}) spectrum for H_2 with $E_{res} = 0.200$ eV and for D_2 with $E_{res} = 0.030$ eV. In a constant residual-energy spectrum the incident electron energy and the scattered electron energy loss are scanned simultaneously with a constant difference E_{res} (from Bose and Linder 1980). For the energy loss peaks numbered 1-5, see Fig. 5.

The intensity I of dipole radiation for a transition $i \rightarrow f$ measured with a polarization-sensitive detector, in the usual notation with ε being the polarization vector and r the transition dipole operator, is given by

$$I = c \sum_{m_f} \langle (i' | \varepsilon \cdot r | f) (f | \varepsilon^* \cdot r | i) \rangle. \quad (1)$$

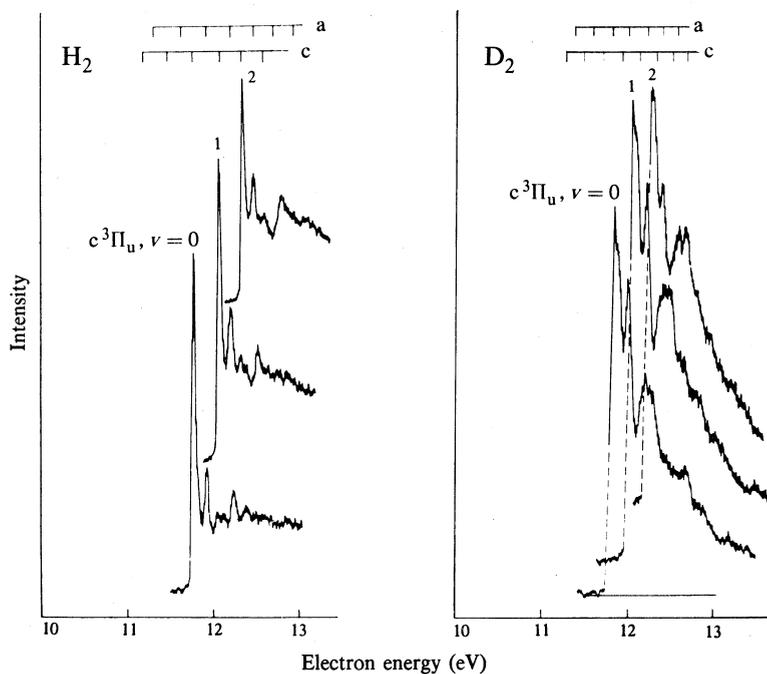


Fig. 3. Typical electron impact excitation functions for the $c^3\Pi_u, v = 0, 1$ and 2 states of H_2 and D_2 (from Bose and Linder 1980).

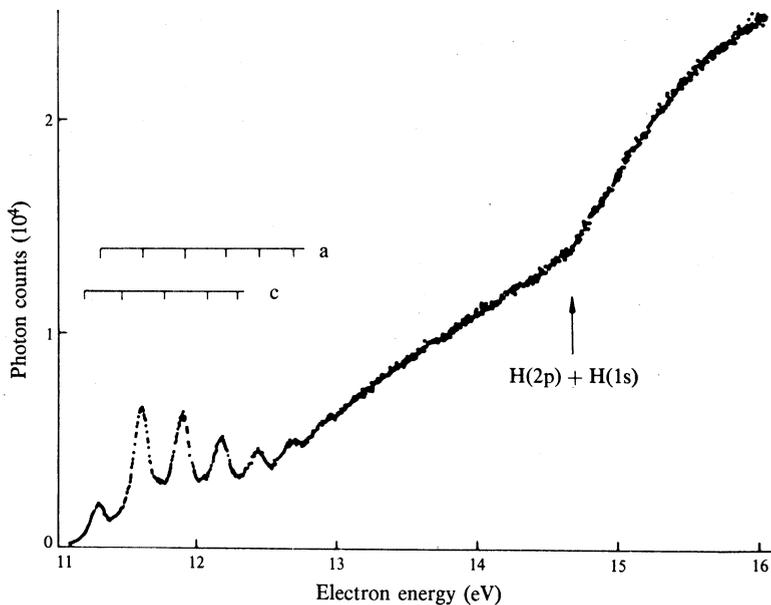


Fig. 4. Optical excitation function for electron impact excitation of H_2 with production of photons in the wavelength range 112–130 nm (from Bose and Linder 1980).

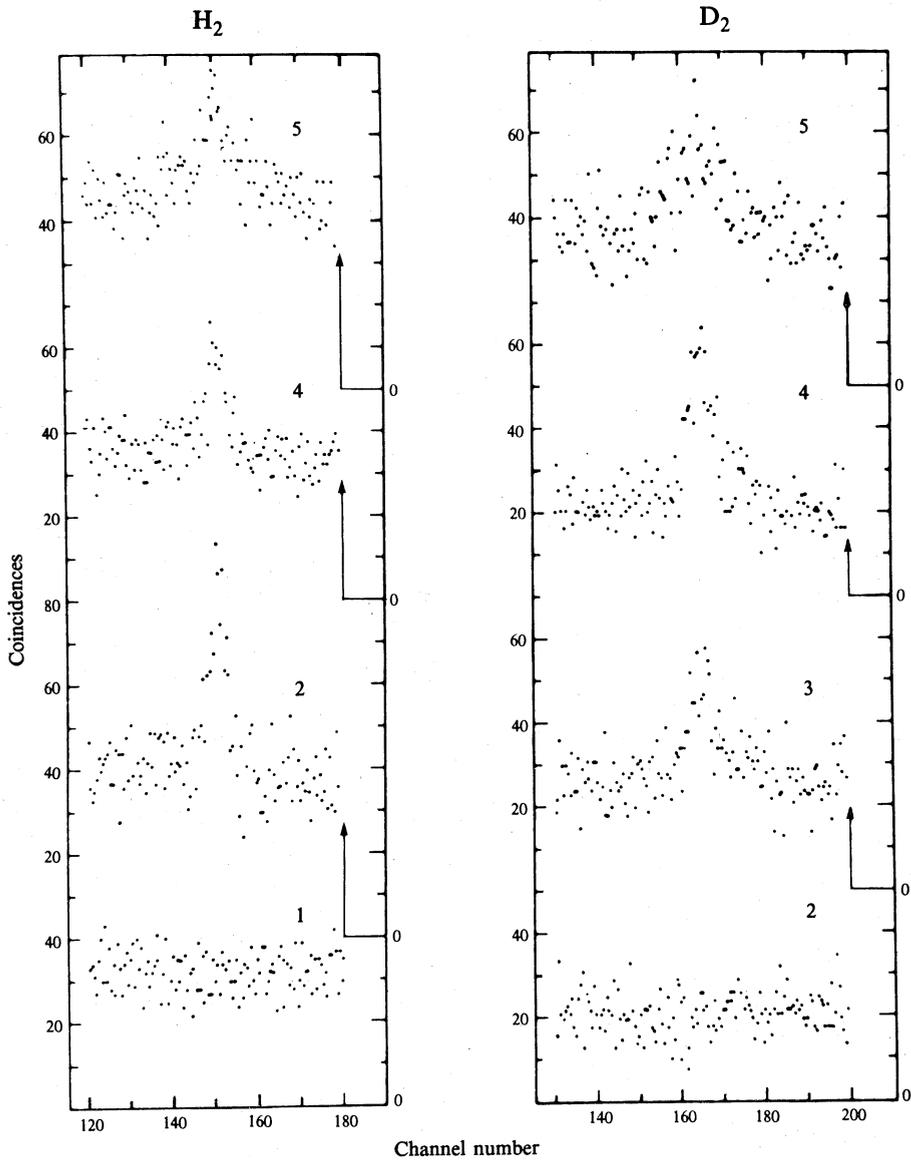


Fig. 5. Coincidences between scattered electrons and radiated photons for H_2 and D_2 shown for the energy loss peaks numbered 1–5 in Fig. 2. True coincidences are seen only for those triplet states lying above the 14.7 eV dissociation limit (for dissociation into $H(1s)$ and $H(2p)$ atoms) (from Bose and Linder 1980).

Then, in a collision reference frame, the angular distribution of photons at time τ emitted in direction \hat{n} is expressed in terms of state multipole angular momentum components by

$$I(\tau) = \sum_{KQ} W(L_r, L) C(K) \left\{ \begin{matrix} 1 & 1 & K \\ L & L & L_r \end{matrix} \right\} D_{KQ}(\hat{n}) G(\tau)_K \langle T(L)_{KQ}^+ \rangle, \quad (2)$$

where $W(L_r, L)$ is proportional to the decay width for producing the detected photons, $C(K)$ is a numerical factor and $D_{KQ}(\hat{n})$ is an element of the rotation matrix to transform the detector frame into the collision frame. Also in equation (2) the perturbation coefficients $G(\tau)_K$ are given by

$$G(\tau)_K = \exp(-\gamma\tau) \bar{G}_K + \frac{\exp(-\gamma\tau)}{2S_1+1} \sum_{J_1 \neq J_1'} (2J_1+1)(2J_1'+1) \left\{ \begin{matrix} L & J_1 & S_1 \\ J_1 & L & K \end{matrix} \right\} \cos\{(E_1 - E_1')\tau/\hbar\},$$

where E_1 and E_1' denote the energies of the coupled states $(LS_1)J_1$ and $(LS_1)J_1'$, and the state multipoles are defined by

$$\langle T(L)_{KQ}^+ \rangle = \sum_{mm'} \rho_{m'm} (-1)^{L-M} (LM'; L-M/KQ), \quad (3)$$

where the density matrix $\rho_{m'm} = W^{-1} \sum a_m^* a_{m'}$ is the weighted average of the various pure atomic states, the a_m being the scattering amplitudes for exciting the magnetic substate m , and $(LM'; L-M/KQ)$ are Clebsch-Gordon coefficients.

Table 1. Relationships between atomic source parameters and experimental observables

Angular momentum components	Source parameter	Multipole moments	Stokes parameters	λ and χ parameters
$\langle 3J_z^2 - J^2 \rangle \{J(J+1)\}^{-1}$	A_0^{col}	$\langle T_0^2 \rangle \{J(J+1)\}^{-1}$	$-\frac{1}{4}\{1 + 3P(0)\}$	$\frac{1}{2}(1 - 3\lambda)$
$\langle J_x J_z + J_z J_x \rangle \{J(J+1)\}^{-1}$	A_{1+}^{col}	$\langle T_{1+}^2 \rangle \{3J(J+1)\}^{-1}$	$-\frac{1}{2}P(\frac{1}{4}\pi)$	$\{\lambda(1-\lambda)\}^{\frac{1}{2}} \cos \chi$
$\langle J_x^2 - J_y^2 \rangle \{J(J+1)\}^{-1}$	A_{2+}^{col}	$\langle T_{2+}^2 \rangle \{3J(J+1)\}^{-1}$	$\frac{1}{4}\{P(0) - 1\}$	$\frac{1}{2}(\lambda - 1)$
$\langle J_y \rangle \{J(J+1)\}^{-1}$	A_{1-}^{col}	$\langle T_{1-}^1 \rangle \{J(J+1)\}^{-1}$	$-\frac{1}{2}P(\text{circ.})$	$-\{\lambda(1-\lambda)\}^{\frac{1}{2}} \sin \chi$

The state multipoles (3) have the simple physical interpretation that $-K \leq Q \leq K$ and $K \neq 0$ defines a polarized state, $K = 1$ defines an oriented state and $K = 2$ defines an aligned state. Excitation of an atom generally leaves it in an anisotropic state, such that the specification of a preferred direction implies that the \mathbf{J} vector is oriented non-randomly ($\langle \mathbf{J} \rangle \neq 0$), which determines the polarization and angular distribution of the decay radiation. The excited state population can be described by expectation values of various combinations of the total angular momentum \mathbf{J} and its components. In the development of this subject, various equivalent parameters have been used, as shown in Table 1.

The λ and χ parameters included in Table 1 were developed by Eminyan *et al.* (1973) for singlet to singlet and $L = 1$ to 0 decay processes, such as $2^1P \rightarrow 1^1S$ transitions in helium where complications due to fine or hyperfine structure do not occur. They have the simple interpretation that λ is the ratio of the differential cross section for excitation of the $M = 0$ sublevel to the total differential cross section, that is,

$$\lambda = |a_0|^2 / (|a_0|^2 + 2|a_1|^2) \quad (4)$$

and χ is the relative phase between the scattering amplitudes a_0 and a_1 .

The study of angular correlations between photons and either scattered electrons, ions or atoms basically concerns the applicability of equation (2) to an excitation collision and the interpretation of the scattering process.

Angular Correlations

The coefficients $G(\tau)_k$ in equation (2) contain the time development of interaction between fine structure of states, hyperfine structure of states with the same L values, as well as the beats between states of different J as, for example, may be coherently excited in atomic hydrogen. The physical process which could occur then is that, during the excitation process, the orbital system acquires a given polarization while the spins remain unpolarized. Because of spin-orbit coupling, there is a transfer of polarization between orbital and spin systems and the spins become polarized with an accompanying loss of polarization of the orbital system, changing $\langle T(L, \tau)_{kQ}^+ \rangle$ as the collision process changes with time. This results in a time modulation of angular distributions and polarization of the emitted radiation. Such experiments have been attempted for beam-foil spectroscopy (Berry 1977) but not yet in particle-photon angular correlations because the factor $\cos\{(E_1 - E'_1)\tau/\hbar\}$ requires appropriate values of E_1 , E'_1 and τ for observable effects. In principle it should also be possible to explore the relationship between time-reversal symmetry and the time evaluation of atomic observables. From time-resolved measurements of the anisotropy and polarization of radiated electric dipole photons, one can expect to construct the atomic observables at the collision time $\tau = 0$.

Present exploration of equation (2) has been limited to s to p state transitions in helium and hydrogen. Extensive measurements of $2^1P \rightarrow 1^1S$ angular correlations in helium have been reported (Arriola *et al.* 1975; Tan *et al.* 1977; Ugbabe *et al.* 1977; Sutcliffe *et al.* 1978; Hollywood *et al.* 1979; Williams 1980) and the theoretical predictions have been discussed by Madison and Shelton (1973), Thomas *et al.* (1974), Roberts (1976), Scott and McDowell (1976) and Blum and Jacobowicz (1978). The excellent experiments of Eminyán *et al.* (1973, 1974, 1975) and Standage and Kleinpoppen (1975) established several basic facts about the excitation process for the $2^1P \rightarrow 1^1S$ and $3^1P \rightarrow 2^1S$ transitions of helium by 80 eV electrons and small scattering angles. The emitted light is completely characterized (Born and Wolf 1970) by the Stokes parameters or equivalently by the polarization density matrix, which specifies the polarization \mathbf{P} and coherence $\boldsymbol{\mu}$ vector properties. Eminyán *et al.* and Standage and Kleinpoppen measured the Stokes parameters to show for the above excitation processes that the 2^1P magnetic sublevels were coherently excited ($\boldsymbol{\mu} = \mathbf{1} = \mathbf{P}$) and that the angular correlations could be analysed in terms of the superposition of pure states. Mirror symmetry in the scattering plane was verified by the reversal of helicity of the circular polarization.

Atomic hydrogen is more interesting. Unfortunately the resolution time of photon detectors is much larger than the spin-orbit precession period and the mean life of the $n = 2$ states so that all measurements in atomic hydrogen will average over the quantum beats. It is not possible, either by electron or photon spectroscopy, to select states with definite angular momentum because of the near degeneracy of levels with the same principal quantum number n but different L . These states are then coherently excited and the detected light is emitted from an ensemble without definite angular momentum. Because the 2s and 2p states have opposite parity, the initial coherence between these states has observational effects only when the atoms

decay in an external field which mixes these states. The theory has been discussed by Blum and Kleinpoppen (1979) and is in advance of experiment for which there are difficulties in obtaining electron angular distributions in the presence of electric fields.

Because no selection is made of initial and final spins there is a summation over the polarizations of the emitted radiation or, in other words, coupling to an unobserved system leads to a depolarization. For the $p \rightarrow s$ transition in H the final state has $L = 0$ so that all transitions from the $L = 1, M_L$ states occur to the same final state so that further depolarization does not occur. Generally both of these factors lead to the degree of coherence $|\mu|$ and the vector polarization $|P|$ being less than unity.

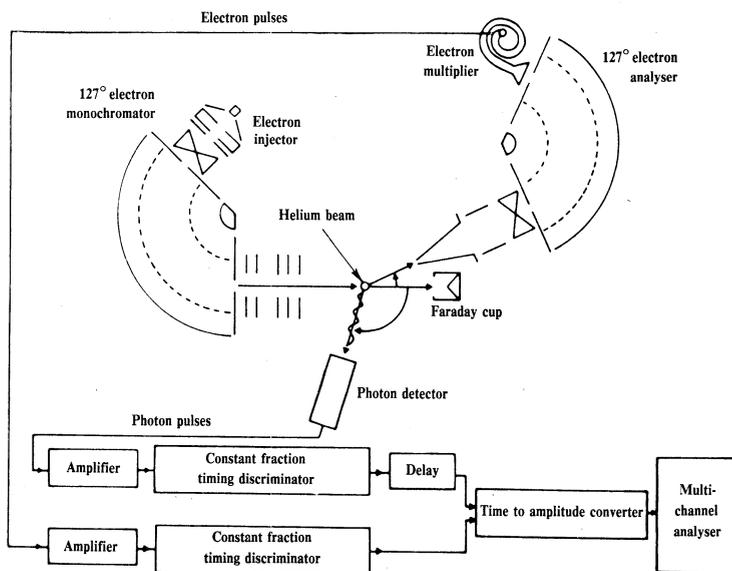


Fig. 6. Schematic diagram of a typical apparatus used in angular correlation studies (from Blum and Kleinpoppen 1979).

Current experiments have been limited to the field-free case and so have observed only the simple 2p dipole photon decay angular correlations. Equation (2) takes the following simple form for the coincidence rate $N(\theta_\gamma)$ between electrons scattered into the direction (θ_e, ϕ_e) following $s \rightarrow p$ excitation with the subsequent emission of a photon in a direction $(\theta_\gamma, \phi_\gamma)$:

$$18 N(\theta_\gamma) = 3\lambda + 11 + 3(1 - 3\lambda)\cos^2\theta_\gamma + 6\sqrt{2} R \sin 2\theta_\gamma \cos(\phi_e - \phi_\gamma) - 3(1 - \lambda)\sin^2\theta_\gamma \cos 2(\phi_e - \phi_\gamma).$$

The first angular correlation measurement in atomic hydrogen was reported by Williams (1975) for 100 eV electron impact excitation of the 2p state. The data were subsequently discussed by Morgan (1976) and Blum and Kleinpoppen (1979). Similar measurements were made by the Flinders group (Dixon *et al.* 1978; Hood *et al.* 1979). All these previous measurements have error bars generally too large

to distinguish between the many theoretical predictions, which is one of the main aims of the measurements. However, the following results (Williams 1980) achieve this aim for a large range of electron momentum transfer.

Let us now collide 54 eV electrons with hydrogen atoms, as shown schematically in Fig. 6, and detect those electrons which have excited the $n = 2$ states and which are scattered through an angle θ_e in coincidence with the emitted 10.2 eV (121.6 nm)

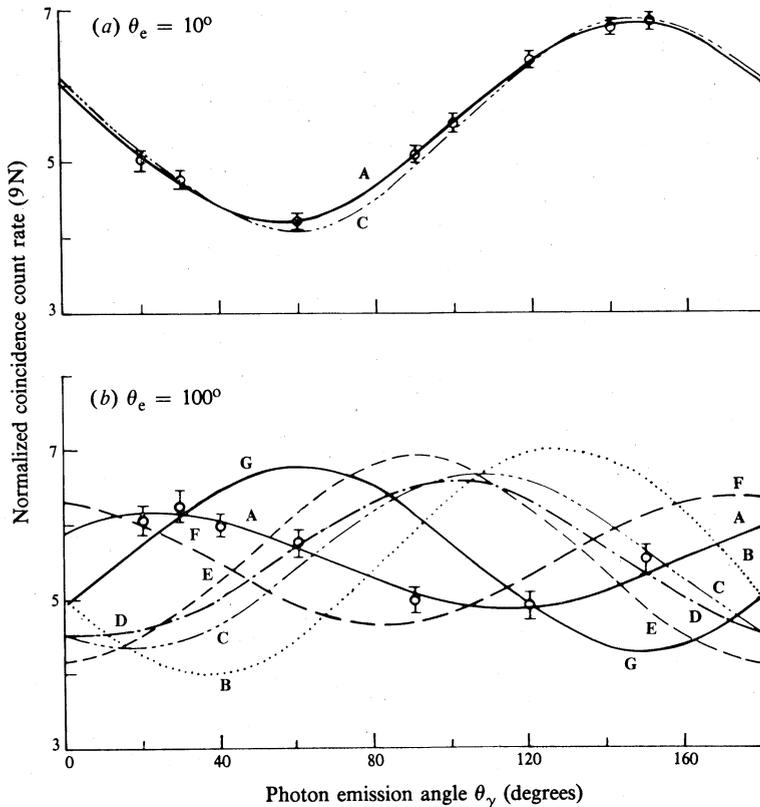


Fig. 7. Electron-photon angular correlations for electron scattering angles θ_e of (a) 10° and (b) 100° and an incident electron energy of 54.4 eV exciting the $2p$ state of atomic hydrogen. The line through the data points (curve A) of Williams (1980) is the unconstrained line of best fit to equation (2). The theoretical curves are: B, first Born approximation; C, classical path T matrix (Roberts 1977); D, distorted-wave Born approximation (Calhoun *et al.* 1977); E, distorted-wave polarized orbital (Morgan and McDowell 1975); F, unrestricted Glauber approximation (Gau and Macek 1974, 1975); G, hybrid close coupling (Kingston *et al.* 1976).

photons at an angle θ_γ . Then the measured coincidence count rate $N(\theta_\gamma)$ for a fixed $\theta_e = 10^\circ$ corresponding to a momentum transfer of $0.39 a_0^{-1}$ over the range of θ_γ from 20° to 150° , as shown in Fig. 7a, is of the form $N(\theta_\gamma) = a + b \cos \alpha$ as given explicitly by equation (2). Fig. 7b shows a similar plot for $\theta_e = 100^\circ$ corresponding to a momentum transfer of $2.9 a_0^{-1}$. The amplitude and phase changes in the sinusoidal function are evident. For the small momentum transfer data the first Born approximation predictions (curve B) are showing differences from the measured data. The measured data for large momentum transfer are clearly not predicted by any theory, including the currently considered best R -matrix theory (Burke and Williams 1978).

By fitting equation (2) to measured data such as in Fig. 7 one can determine the parameters λ and R for each θ_e . Fig. 8 shows the data of Williams (1980) to be in agreement with those of Weigold *et al.* (1980). The difficulty of the measurements is determined by the differential 2p state excitation cross section which decreases by more than two orders of magnitude as θ_e increases from 10° to 140° . At small θ_e the close coupling theory of Kingston *et al.* (1976; curve G) predicts values of λ that are within the 70% confidence limits of the measured values, but, as indicated in Fig. 7b for $\theta_e = 100^\circ$, at large scattering angles none of the theoretical methods predict the measured data.

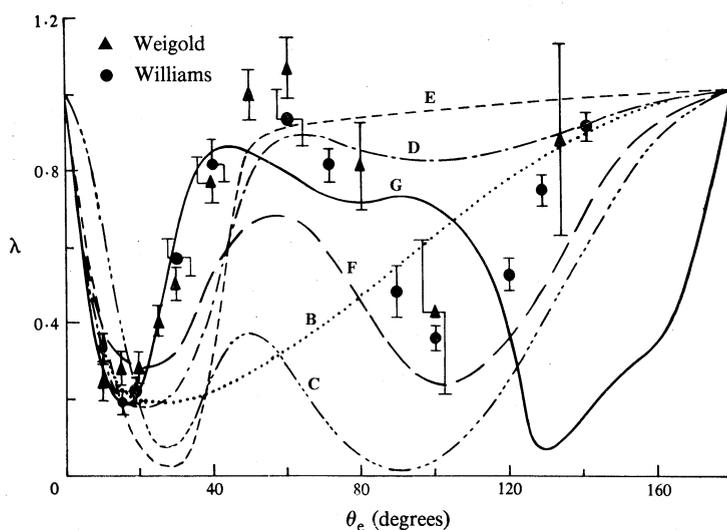


Fig. 8. Plot of the parameter λ as a function of the electron scattering angle θ_e for an incident electron energy of 54.4 eV. The experimental data of Weigold *et al.* (1980) and Williams (1980) are compared with the theoretical curves B-G, as defined in Fig. 7.

With the simple definition of equation (4) for λ , the physical interpretation can be given that, as λ changes from unity to zero, the population of the magnetic sublevels shifts from the $M_L = 0$ to the $M_L = \pm 1$ levels. Equation (2) simplifies for $\phi_e - \phi_\gamma = \frac{1}{2}\pi$ and $\theta_\gamma = \frac{1}{2}\pi$ to give $N(\theta_\gamma) = 7/9$, independent of both λ and R . In this case it is readily shown that both the ratio $\sigma(2s)/\sigma(2p)$ and $\sigma(2p)$ may be independently measured from measurements of the coincidence count rate. Then the values of λ and $\sigma(2p)$ continue to give absolute values of σ_0 and σ_1 , which are shown in Fig. 9. There appear to be two interactions dominating the population of the $|M_L| = 1$ states, one causing the strong forward peak and a second causing a four orders-of-magnitude weaker and broader peak near $\theta_e = 90^\circ$. Similarly there appear to be two $|M_L| = 0$ population mechanisms, one causing a strong forward peak and a second causing perhaps an isotropic distribution above $\theta_e = 100^\circ$.

In summary electron scattering from atomic hydrogen is a simple three-body problem and yet, for even the excitation of the lowest $n = 2$ state, the best theory to date cannot describe measurements of the alignment parameter at large momentum transfers. State-of-the-art experimental techniques now permit measurements of the individual L and M_L level differential cross sections.

Related Studies

The previous section has described only directional correlation measurements; however, McConkey and Malcolm (1979) have explored polarization correlation techniques, for which a schematic representation is given in Fig. 10. The two methods give similar information, as discussed, for example, by Steffen and Alder (1975). Difficulties are involved in applying the polarization correlation technique to Lyman- α radiation because of the lack of an efficient circular polarizer. However, only this technique gives the helicity of the radiation which is required for a complete specification of the excited state density matrix.

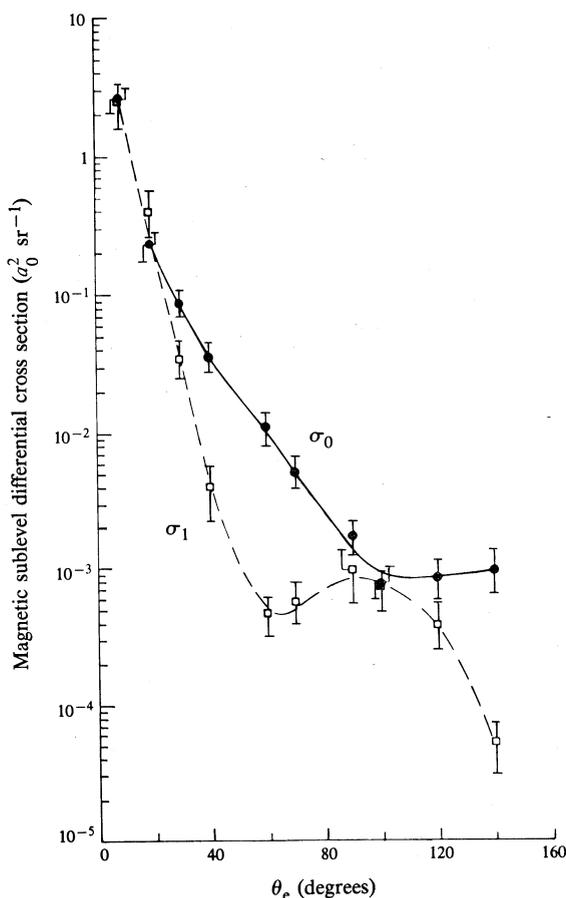


Fig. 9. Differential cross sections (from Williams 1980) for excitation of the individual magnetic sublevels $|M_L| = 0$ and 1 of the 2p state of atomic hydrogen, shown as a function of the electron scattering angle θ_e for an incident electron energy of 54.4 eV. The data have been derived from measurements of $\sigma(2p)$ and the angular correlation parameter λ .

A further disadvantage of the coincidence method described above is that it is not possible with this method to obtain information on state multipoles $\langle T^K \rangle$ with $K > 2$ (Blum and Kleinpoppen 1979). This limitation can be overcome in principle using the inverse of the electron-photon coincidence method, as pointed out by

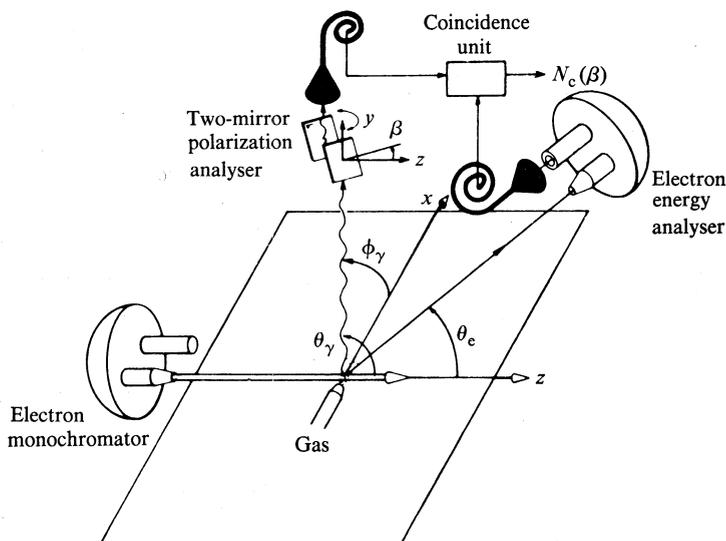


Fig. 10. Schematic diagram of a typical apparatus used in polarization correlation studies (from Malcolm and McConkey 1979).

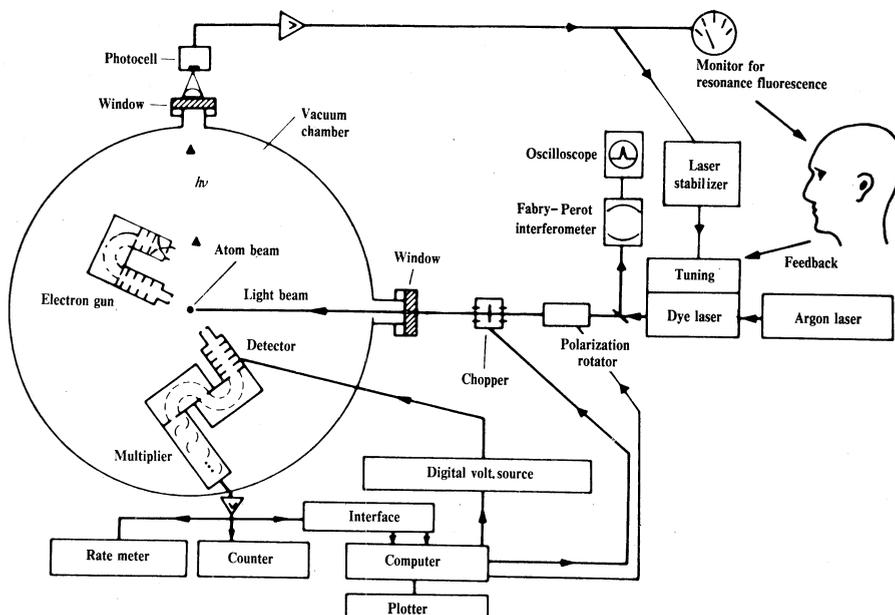


Fig. 11. Schematic diagram of an apparatus used for studies of electron scattering from laser-excited atoms (from Hertel and Stoll 1977).

Hertel and Stoll (1977). A schematic representation of their approach is shown in Fig. 11. An atom is prepared in a well-defined magnetic substate of an excited state by photon absorption from a laser beam. Electrons scattered from this excited state are then detected. The same multipole components are involved as in the above time-inverse scattering process, but the laser selection of the magnetic substates in principle permits the determination of the higher order ($K > 2$) multipole moments. Application of the method has so far been limited to sodium atoms because of the lack of suitable lasers.

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