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(e, 2e) QUASI-FREE ELECTRON SCATTERING ON BOUND
ELECTRON. COINCIDENCE MEASUREMENTS OF SCATTERED
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R. Camilloni^(x), A. Giardini Guidoni, G. Stefani^(o) and R. Tiribelli: (e, 2e)
QUASI-FREE ELECTRON SCATTERING ON BOUND ELECTRON. COINCIDENCE MEASUREMENTS OF SCATTERED AND EMITTED ELECTRONS IN CARBON. -

ABSTRACT. -

Momentum distribution of bound electrons belonging to K and L shells in Carbon have been separately measured. Data obtained from the angular distribution of coincidences between the two electrons outcoming from a quasi-free (e, 2e) process have been compared with theoretical previsions.

INTRODUCTION. -

Recently there has been a revival in the interest of actually measuring electron momentum distribution (e. m. d.) in atoms molecules and crystals. The techniques used in order to obtain these distributions are: Compton scattering of X rays⁽¹⁾, quasi-free electron scattering at large angles⁽²⁾ and positron annihilation⁽³⁾. In the experiments published up to now only one of the three reaction products has been detected. This fact constitutes an important limitation, because it makes impossible to resolve e. m. d. 's belonging to electron in different bound states. Despite of this limitation interesting information has been obtained, especially from the study of the Compton line broadening. They have allowed some critical comparisons with the computed electron states in atoms and crystals. Even when the initial state of the bound electron is defined, as for instance by taking the scattered X ray in coincidence with a fluorescence photon⁽⁴⁾, the Compton profile does not give direct information on the e. m. d. function, because it is connected only with a suitable integral of this function. To overcome these difficulties it seems very

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promising to measure the angular correlation of the scattered and ejected electrons taken in coincidence. The process has to be a "quasi-free" scattering and the energies and momenta of the particles must be measured very carefully. In this way a direct measurement of e. m. d., and therefore of the wave function itself, of a well defined initial state can be achieved. Of course the improvement of information goes together with an increase of experimental difficulties, due to the smaller value of the cross sections involved and to much more stringent experimental requirements.

Since many years theoretical works are published^(5, 6) suggesting the use of a coincidence technique in measuring e. m. d. in atoms, molecules and solids. Experimental results are still lacking. Only Amaldi et al.⁽⁸⁾ demonstrated the feasibility of these (e, 2e) experiments⁽⁹⁾. In their papers an apparatus has been described whose resolving power is such to distinguish (e, 2e) events outcoming from external or internal Carbon shells. Preliminary measurements were performed in Formvar targets and angular distributions for coincidences events presented.

In the present paper we discuss the results on the momentum distribution of electron states, obtained by using as targets evaporated Carbon films (Yissum Research Development Comp. Jerusalem, Israel), put in a modified and improved version of the apparatus used in ref. (8).

1. - QUASI-ELASTIC SCATTERING. KINEMATICS OF THE PROCESS. -

Let us write the (e, 2e) process as:



where A is the target atom and R is the residual ion. The conditions which must be satisfied to insure that the observed coincidence events can be considered as due to a quasi-free electron scattering have been already discussed⁽¹⁰⁾. In this paper we will limit ourselves to the enunciation of these conditions, which are also necessary to guarantee that the collision cross section can be calculated in the framework of the Impulse Approximation (I. A.).

They are essentially the following:

- a) the scattering event has to be localized in a volume which is much smaller than the atomic dimensions;
- b) the incoming and final electron energies have to be much greater than the binding energy of the target electron.

When these conditions are satisfied, the single particle character of the wavefunction of target electron is emphasized and the remainder of the system (nucleus + residual electron cloud) acts only as a spectator. Essentially it produces the force field which determines the initial state of the target electron, but does not affect the electron-electron interaction.

Conditions a) and b) are equivalent to:

$$(1.1) \quad \left| \Delta \underline{p} \right| = \left| \underline{p}_0 - \underline{p}_s \right| \gg \frac{\hbar}{\Delta r}, \quad E_0 \gg B_{nl}, \quad E_s, E_e \gg B_{nl}$$

where E_0 and \underline{p}_0 are respectively the energy and the momentum of the incident electron, $E_s, \underline{p}_s, E_e, \underline{p}_e$ the energies and the momenta of the scattered and ejected electrons, B_{nl} is the binding energy and Δr , the localization of the event, is of the order of a Bohr radius.

The kinematical quantities are related by energy and momentum conservation laws:

$$(1.2) \quad \underline{p}_0 = \underline{p}_s + \underline{p}_e + \underline{p}_R$$

$$(1.3) \quad E_0 - B_{nl} = E_s + E_e + E_R$$

In a knock-out reaction the momentum of the non interacting residual ion R is conserved. Since \underline{q} is the electron momentum in the A system initially at rest, the following relationship holds

$$(1.4) \quad \underline{p}_R + \underline{q} = 0$$

and the equations (1.2 and 1.3) become

$$(1.5) \quad \underline{p}_0 + \underline{q} = \underline{p}_s + \underline{p}_e$$

$$(1.6) \quad p_0^2 - \alpha^2 = p_s^2 + p_e^2$$

where $\alpha^2 = 2m_e B_{nl}$, and the recoil energy E_R has been neglected. In our hypotheses equations (1.5 and 1.6) describe a kinematics very similar to the scattering of two free electrons, one of which at rest. In these conditions the two final particles are emitted at a total angle of 90° in the laboratory system. When the target electrons have an initial momentum distribution this complete correlation is partially destroyed.

The case of completely symmetrical coplanar scattering implies:

$$(1.7) \quad p_s = p_e = (p_0^2 - \alpha_{nl}^2)^{1/2} \frac{1}{\sqrt{2}}$$

$$\theta_s = \theta_e$$

so that by varying the angles near the value

$$(1.8) \quad \theta_0 = \frac{1}{2} \arccos \frac{B_{nl}}{(E_0 - B_{nl})}$$

4.

which corresponds to the value $\underline{q} = 0$, and keeping the final momenta of the electrons fixed, only momenta \underline{q} of the initial electron parallel to \underline{p}_0 are explored (Fig. 1).

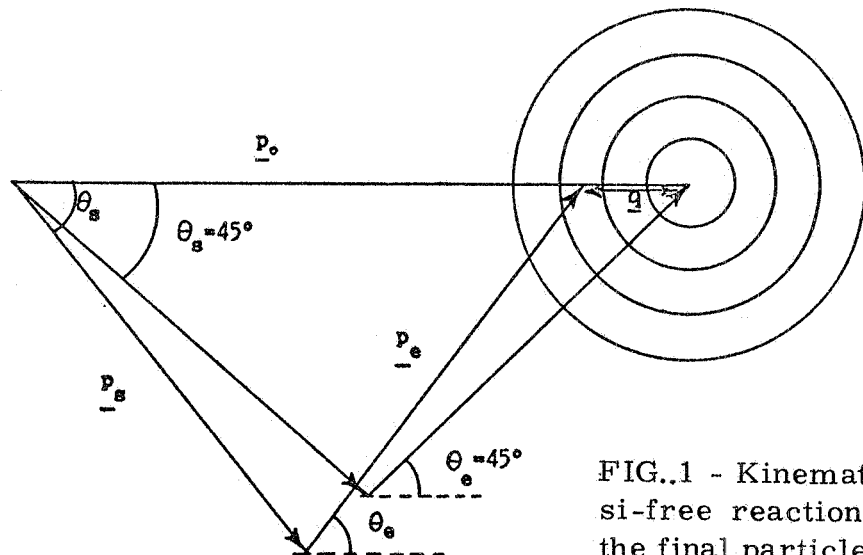


FIG.1 - Kinematical diagram of a quasi-free reaction at fixed energies of the final particles.

For spherical distributions this kinematical condition does not imply limitations on \underline{q} . The measured \underline{q} values are:

$$(1.9) \quad \underline{q} = (2 p_s \cos \theta_s / 2 - p_0) \hat{p}_0$$

2. - (e, 2e) CROSS SECTION. -

The hypotheses a) and b) of section 1 guarantee that the I. A. can be applied, as it is proved in reference (8). In the same work a critical comparison of the Born Approximation and of the I. A. is carried out, which shows that in atomic physics the two approximations are equivalent, owing to the weakness of interaction (cfr. also Glassgold etc. ref. (5)).

Evaluations of the (e, 2e) cross sections appear in the quoted theoretical papers^(5 ÷ 7). In ref. (10) it is pointed out that they differ by numerical factors. The correct expression for the cross section differential in the energy of one of the final electrons and in the two solid angles, can be written as follows⁽¹⁰⁾:

$$(2.1) \quad \frac{d^5 \sigma}{d\Omega_s d\Omega_e dE_s} = \frac{m p_e}{\hbar^3} \left(\frac{d^2 \sigma}{d\Omega_{s f. e.}} \right) \varrho_{nl}(\underline{q})$$

In this expression

$$(2.2) \quad \left(\frac{d^2 \sigma}{d\Omega_s f. e.} \right) = 4 m^2 e^4 \frac{P_s}{p_o} \left(\frac{1}{P^4} + \frac{1}{S^4} - \frac{\cos(\eta_p - \eta_s)}{P^2 S^2} \right)$$

is the free electron scattering cross section, where

$$\underline{P} = \underline{p}_o - \underline{p}_s \quad , \quad \underline{S} = \underline{p}_o - \underline{p}_e \quad , \quad \eta_p - \eta_s = \frac{2 e^2 m_e}{\hbar |\underline{p}_e - \underline{q}|} \ln \frac{S}{P}$$

and $e_{nl}(\underline{q})$ is the squared Fourier transform of the overlap integral of the final ion and initial atom wave functions. For atoms, in the Self Consistent Field approximation (S. C. F.), the form factor becomes:

$$(2.3) \quad e_{nl}(\underline{q}) = \frac{1}{2l+1} \sum_{m=-l}^{+l} \frac{1}{(2\pi)^3} \left| \int e^{i \frac{\underline{q}}{\hbar} \cdot \underline{r}} \varphi_{nlm}(\underline{r}) d\underline{r} \right|^2$$

where the sum indicates average over the spins. The (nl) state and \underline{q} are obviously identified by energy and momenta conservation laws (1.5 and 1.6). In Fig. 2 we report calculated cross sections for some states of Silver in the kinematical and experimental conditions of our apparatus (cfr. section 4) (symmetrical coplanar scattering). The atomic wavefunctions were obtained from the M. B. S. by Clementi⁽¹¹⁾.

For crystals the form factor is very sensitive to the different models presently adopted to account for the band states.

This subject was discussed in a very recent paper (Levin et al.⁽¹²⁾) and previously in a thesis work⁽¹³⁾. As a result, it seems that one may achieve precise informations on the states of the electrons in the bands. From ref. (13) two cases are reviewed in order to clarify our previous statement on the form factors.

a) - Free electron approximation. -

The wavefunction is described by a plane wave

$$\psi(\underline{r}) = \frac{1}{\sqrt{V}} e^{i \underline{K} \cdot \underline{r}}$$

where V is the volume of the crystal and the wavevector \underline{K} is related to the momentum \underline{q} of the electron as follows:

$$\underline{K} = \underline{q}/\hbar$$

The cross section (2.1.) becomes

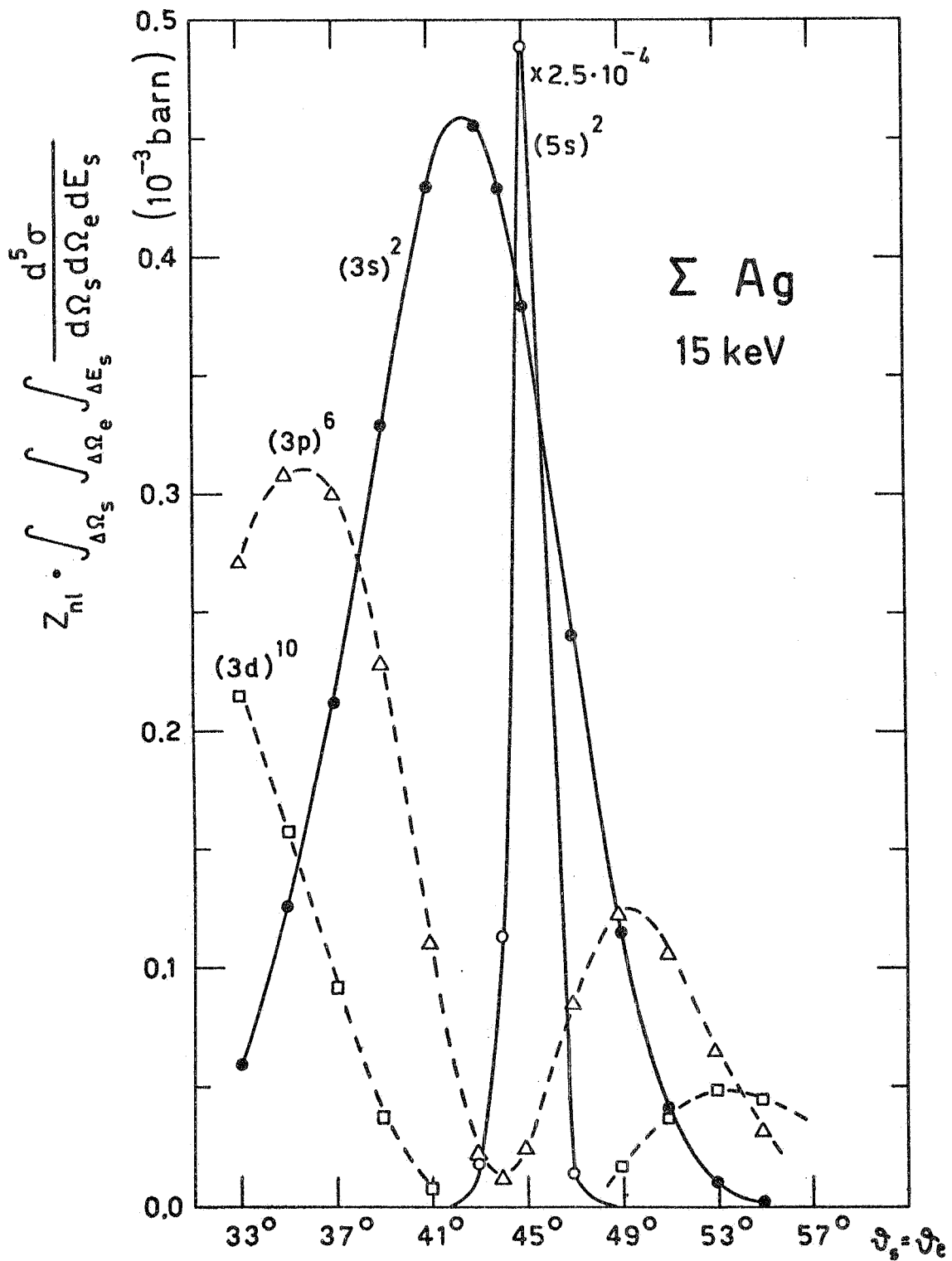


FIG. 2 - Calculated cross sections for Silver in symmetrical scattering. Wave functions are obtained by M.B.S. method. Integration ranges are relative to our experimental apparatus.

$$(2.4) \quad \frac{d^5 \sigma}{d\Omega_e d\Omega_s dE_s} = \frac{m p_e}{\hbar^3} \left(\frac{d^2 \sigma}{d\Omega_{s f. e.}} \right) G_{fi} \left[1 - \frac{\underline{K} \cdot \nabla_{\underline{K}} \varepsilon_1(\underline{K})}{2 E_e} \right]^{-1}$$

where $\varepsilon_1(\underline{K})$ is the energy eigenvalue of the state with wavevector \underline{K} , in the band 1.

The form factor G_{fi} reduces to a δ -function of the momentum:

$$G_{fi} = \delta(\underline{q} - \hbar \underline{K})$$

The term in square brackets originates from the energy conservation law and in the case of the atoms reduced to unity, because the eigenvalue of the energy does not depend on the momentum.

b) Strong coupling approximation. -

$$\psi_{1, \underline{K}}(\underline{r}) = \frac{1}{\sqrt{N}} e^{i \underline{K} \cdot \underline{R}} \varphi_1(\underline{r} - \underline{R})$$

where N are the atoms having lattice coordinates \underline{R} ; \underline{K} is the so-called quasi-momentum of the electron. The form factor becomes in this case:

$$G_{fi} = \left| \varphi_1(\underline{q}) \right|^2 \delta(\underline{q}/\hbar - \underline{K} - \underline{B})$$

where $\varphi_1(\underline{q})$ is the atomic function generating the band, and \underline{B} is a reciprocal lattice vector.

The cross section in this case is non zero for all values of $(\underline{K} + \underline{B})$ which reduce to zero the argument of the δ -function.

The other approximations treated in the ref. (13), as the O. P. W. method or the case of polyatomic crystals, are not reported here for sake of brevity, but they plentifully confirm the sensitivity of the form factor to the shape of the wavefunction.

3. - EXPERIMENTAL LIMITATIONS. -

In this section, the main difficulties which can be encountered in performing (e, 2e) experiments are briefly recalled, and the limitations involved are shown.

The first difficulty is connected to the presence of the accidental coincidence background which can limit the number of the electronic states

to be studied^(x). When this ratio becomes too low, it is no longer possible to distinguish the true events from the background fluctuations. These conditions are encountered in the study of internal states of heavy elements, since to larger average momenta corresponds a flattened shape of distribution and since the contribution of different states increases with Z .

The second difficulty concerns the techniques required to obtain reliable data. Indeed the energy and angular correlations of the electrons might be destroyed by energy losses. This implies that, while there are no limitations in the study of gases even at not very high incident electron energy, experimental difficulties arise for solid targets. It is well known that the minimal thickness of a self supporting film, few μm . in diameter, cannot practically be reduced to less than few hundreds \AA ⁽¹⁵⁾. However these values are still too large for energy losses being completely neglected at incident energies of about 10 KeV also for energy resolutions of about one hundred eV. On the other side an energy increase limits the spread of coincidence events due to the momentum distribution in external states to a very small angular range ($\Delta\theta \simeq 1^\circ$), and an apparatus of very good angular resolution is needed. In conclusion, for a given target thickness, the choice of the beam energy has to balance two effects: the energy losses, which have to be minimized, and the shrinkage of the angular breadth.

A possible different approach can be found in the use of grazing angles for the incoming momentum p_0 .

However it should be borne in mind that in this kinematical condition q values near zero are impossible to measure.

Let us consider the case of electrons incident on a crystal at an angle $\phi_{\text{inc}} \neq 0$ respect to the surface. By collecting the two electrons emerging at an angle ϕ_{ex} , a component q_{\perp} of the momentum of the bound electron is selected by the equations (1.5 and 1.6), along the direction perpendicular to the surface, Fig. 3. When the incident energy is $E_0 \simeq 3 \text{ KeV}$ and the angles are $\phi_{\text{in}} = \phi_{\text{ex}} \simeq 10^{-2}$ rad, the value of q_{\perp} is about 0.5 KeV/c.

Because in crystals the main interest is to determine the momentum distribution in valence and conduction bands, for which the average momentum is quite small, (typical values of the momentum $\hbar K$ at the boundary of the first Brillouin zone are 1-2 KeV/c), a not negligible part of the e.m.d. function cannot be measured, and this experimental arrangement appears very limiting.

(x) - This subject has been more widely treated in ref. (10, 15) where the character of "unavoidable" coincidences is pointed out and the ratio $N_{\text{nl}}/N_{\text{acc}}$ of the true events rate (N_{nl}) to the accidental rate (N_{acc}) is estimated.

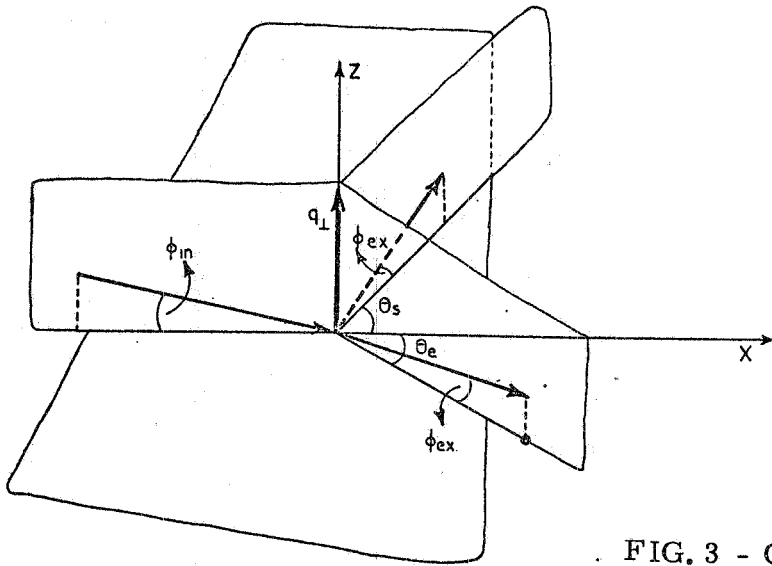


FIG. 3 - Grazing angle kinematics.

4. - THE APPARATUS. -

The block diagram of the apparatus is shown in Fig. 4. The apparatus consists of a scattering chamber 35 cm in diameter, pumped by an oil diffusion pump to 10^{-6} torr, which contains an electron gun, the target, a Faraday cup monitoring the transmitted intensity of the current, and two twin electrostatic deflectors followed by two channeltron detectors.

The electron gun previously described in ref. (10) furnishes a quite collimated beam (angular spread $\Delta\theta \approx 0.5^\circ$) of small spot dimensions ($\phi \approx 0.5$ mm). The position of the beam is mechanically adjustable under vacuum into a range of about 10 mm of diameter. Maximum current intensity is about 10^{-6} A. The beam energy can be varied from 4 to 20 KeV without affecting its characteristics. The electrostatic deflectors, the same previously used⁽⁸⁾, have a resolving power $\Delta E/E = \pm 1\%$ and an accepted angle $\Delta\Omega \approx 7 \cdot 10^{-4}$ ster. Two couples of cylindrical electrostatic plates, placed at the entrance of each analyzer, partly correct for fringing field effects and optimize the angular symmetry of the deflectors.

The data acquisition system is essentially the same of ref. (8). Coincidence resolving time for saturated signals of the channeltrons is 5 nsec.

5. - EXPERIMENTAL RESULTS. -

Coincidence measurements have been performed in symmetrical coplanar scattering conditions. Scattering angles are measured by a direct method, independent from the mechanical calibration. It is well known, indeed, that the quasi-elastic peak position, in the single channel energy spectra, is found at an energy value E_{qe} , which is related to the scattering angle θ by (2):

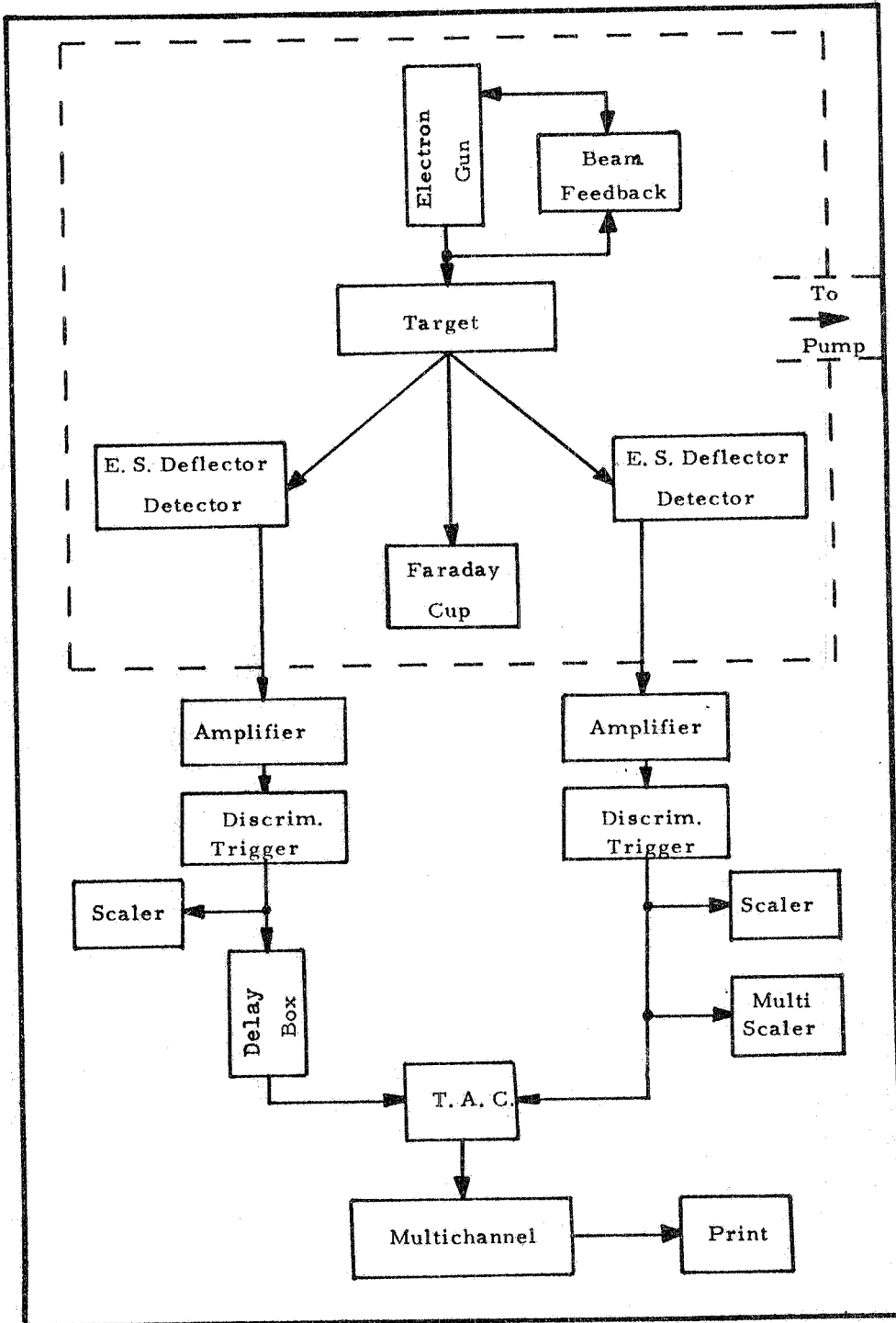


FIG. 4 - Block diagram of experimental set-up.

$$E_{qe}/E_0 = \cos^2 \theta$$

where E_0 is the energy of the elastic peak. In Fig. 5 some typical spectra

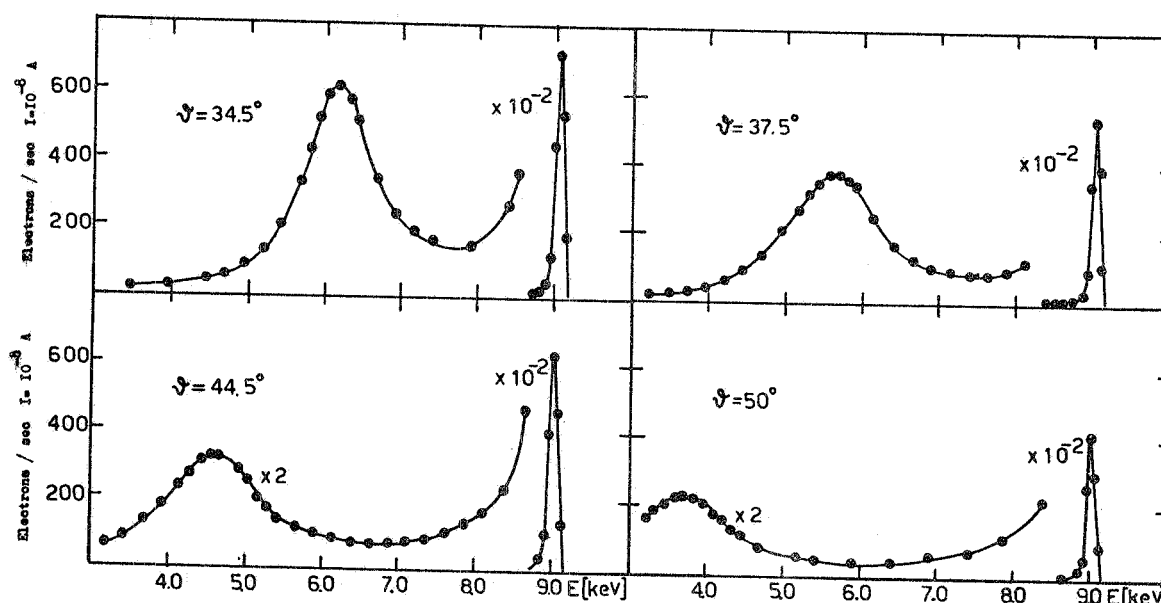


FIG. 5 - Energy spectra of 9 KeV electrons scattered through different angles by Formvar targets.

are reported, for various angles and Formvar targets (obtained by a solution 0.1% molar in dichloroethane). Such spectra have been recorded for both the final electrons, before every run at fixed angle. The uncertainty in the determination of quasi-elastic peak position causes an error $\Delta\theta \approx \pm 0.5^\circ$, smaller than the mechanical reliability.

Targets are formed of evaporated Carbon films, holded by an electroformed mesh (Ni 82% transmission). Damage effects are avoided by shifting the holder under the beam about every 10 or 15 min. Typical values for incident beam are: Intensity $3.5 \cdot 10^{-8}$ A; Energy 9.0 KeV.

Figure 6, 7, 8 show some coincidence energy spectra, relative to various angles and target thicknesses. Two peaks can be seen, whose energy difference is about 300 eV. They are assigned to electrons extracted from the K and L shells, of Carbon, whose separation energy is about 278 eV. The change of the relative intensities of the two peaks with angle is in agreement with the narrower angular distribution of the L electrons carrying lower momenta.

The experimental points reported in the figures 6, 7, 9, 10 are the average of N consecutive (variable from 3 to 5) determinations on different target points, normalized to the elastic peak value.

The angular distribution of the true coincidence rate for 1s state is shown in Fig. 9. For comparison curves are reported as obtained from

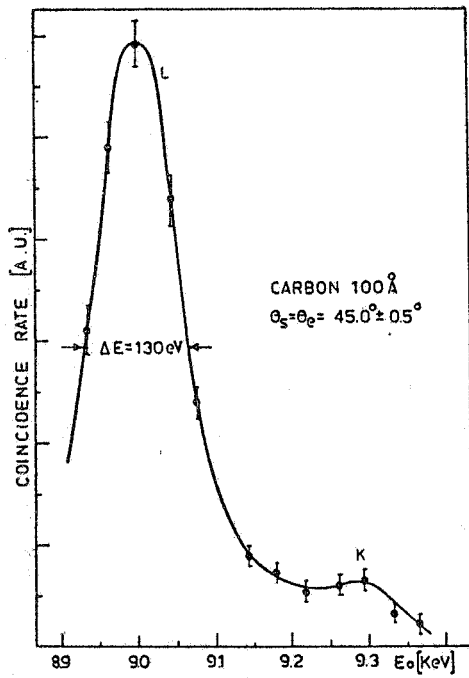


FIG. 6 - Coincidences energy spectrum for 100 Å thin Carbon. Angular conditions : $\theta_s = \theta_e = 45.0^\circ \pm 0.5^\circ$.

FIG. 7 - Coincidences energy spectrum for 250 Å thin Carbon. Angular conditions: $\theta_s = \theta_e = 43.5^\circ \pm 0.5^\circ$.

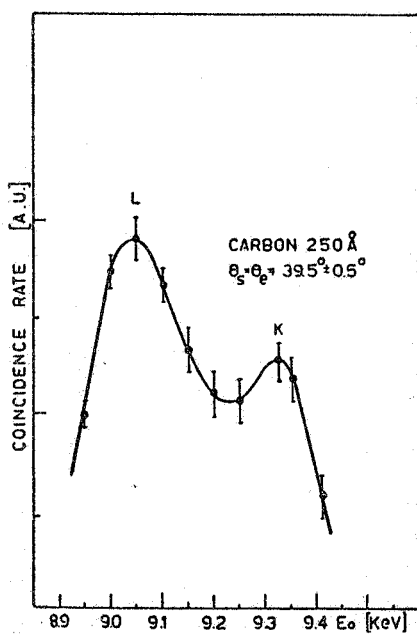
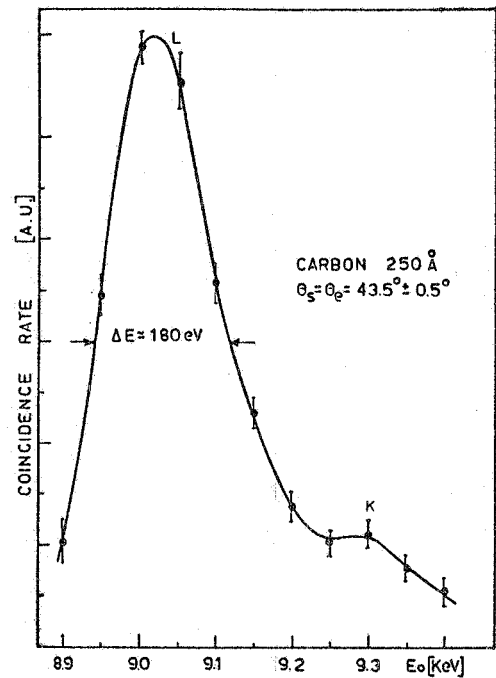


FIG. 8 - Coincidences energy spectrum for 250 Å thin Carbon. Angular conditions : $\theta_s = \theta_e = 39.5^\circ \pm 0.5^\circ$.

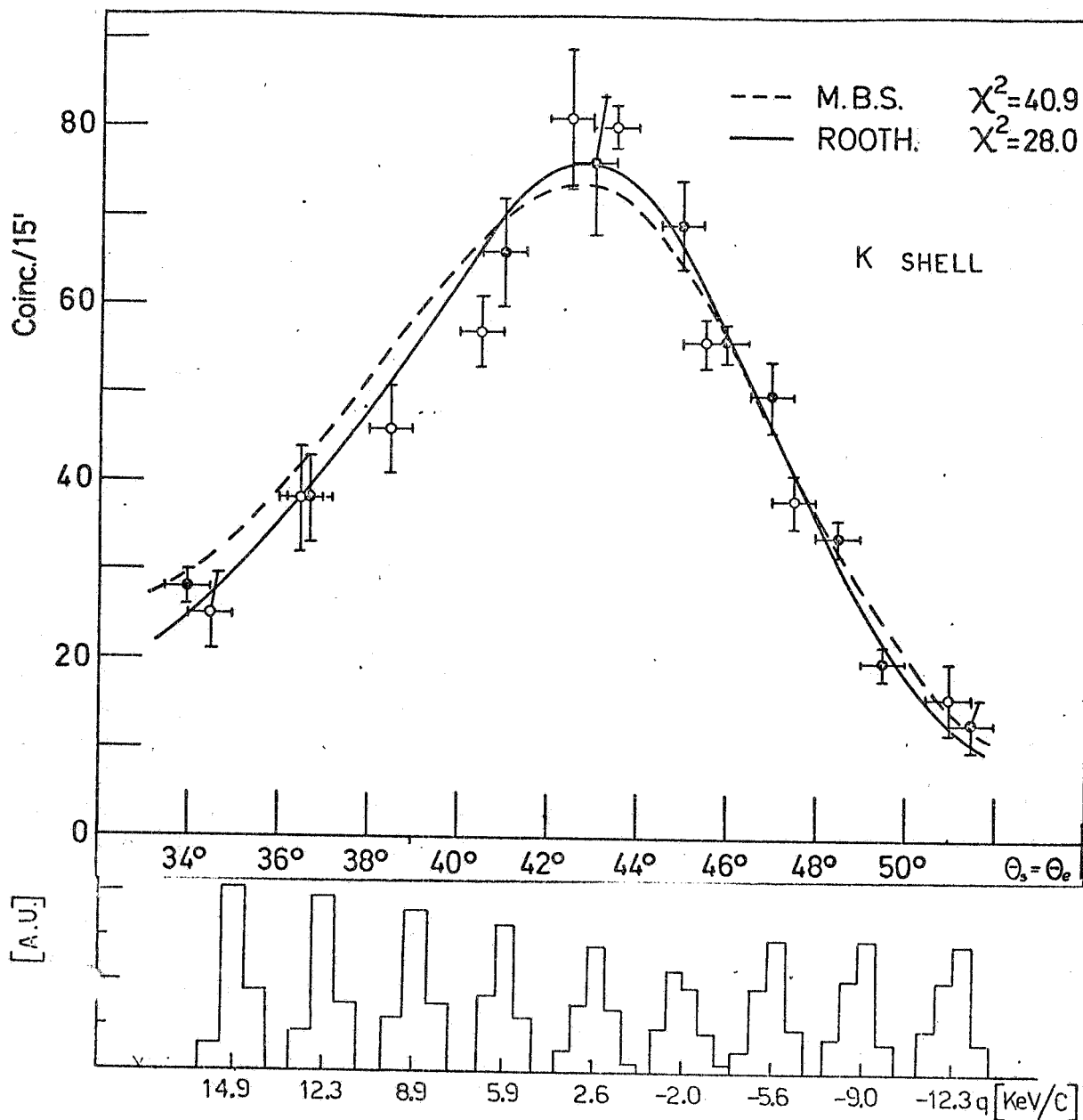


FIG. 9 - Angular distribution of the coincidence rate for the 1s state of Carbon. Experimental conditions: beam energy $E_0 = 9.3$ KeV; final energies of the electrons $E_s = E_e = 4.5$ KeV; current intensity $3.5 \cdot 10^{-8}$ A. The data belong to two different series of measurements on two 250 Å thick Carbon films. The correspondence between scattering angle and initial momentum of the atomic electron is also given. The dashed and continuous curves are the predictions obtained with M. B. S. and Roothaan wave functions.

Roothaan and M. B. S. wavefunctions (full curve and dashed curve respectively)⁽¹¹⁾.

The (e, 2e) cross section (2. 1) has been integrated over the angular and energetic experimental resolutions by means of a computer program, which properly accounts for the transmission functions of the analyzers. For some angles momenta sampled by our apparatus along the incident beam direction are reported.

The maximum of the curves is not found at the momentum $q = 0$, because of the factor $1/\sin^4\theta$ arising from the free (e, e) cross section (2. 2).

Analogous data concerning the external L shell are reported in Fig. 10. They are relative to two different target thicknesses and have been normalized to the same area.

6. - DISCUSSION. -

Before interpreting the experimental curves reported in Fig. 9 and Fig. 10, it is necessary to discuss the problem of energy losses suffered by electrons of a few KeV energy traveling through a film few hundreds Å thick. One of us⁽¹³⁾ treated in detail how these interactions can destroy informations obtainable from (e, 2e) processes. This happens when direction changes due to the collisions are large compared to the resolution required for mapping the angular distribution, or when the energy shift is such that events belonging to different states overlap. In intermediate cases the only effect is a lowering of counting rate.

For thin Carbon targets data on energy losses for various incident electrons energies are available⁽¹⁶⁾. By extrapolating these data, according to the cross section reported by Raether⁽¹⁷⁾, the mean free paths for electrons of 9 and 4.5 KeV have been computed to be $\lambda = 105 \pm 10$ and $\lambda = 60 \pm 5$ Å respectively. For each scattering event in glassy Carbon an energy loss of 20 eV⁽¹⁸⁾ and an average angular spread $\Delta\theta \ll 10^{-3}$ rad⁽¹⁹⁾ can be assumed. By estimating the probability of an electron being scattered n times by using a Poisson distribution, we can compare the angular and energetic shifts suffered by the incident and final electrons with our experimental conditions (accepted angle $\Delta\theta \simeq \pm 8 \cdot 10^{-3}$ rad, energy resolution for each channel $\Delta E_{1/2} = \pm 45$ eV at half height).

The calculation indicates that a not great effect of losses should be observed in a target ≤ 100 Å thick, while a remarkable lowering of counting rate is expected for 250 Å. In agreement with these predictions, the full width for the coincidence peak of L electrons in Fig. 6 is found $\Delta E_{1/2} = 130$ eV. This value is expected by combining two Gaussian distributions, $\pm 1\%$ wide (± 45 eV), and imposing that the coincident electrons conserve the energy, as in equation (1. 6). The curve of Fig. 7, relative to a film 250 Å thick, shows a greater full width $\Delta E_{1/2} = 180$ eV. Due to this fact the absolute values of the L and K maxima are lower than expected.

Notwithstanding, counts belonging to the different shells do not overlap at the maxima of the two peaks and the angular deviation due to the losses is still negligible (10^{-3} rad).

Therefore, it can be correctly assumed that the energy losses reduce only the absolute value of the maxima of the peaks, without affecting the shape of angular distributions. It is so possible to interpret them in terms of electron momentum density, i. e. in terms of the wavefunctions Fourier transform.

The best fit of the data for 1s state, Fig. 9, is obtained by minimizing the χ^2 function with unknown apparatus efficiency. The χ^2 test gives a confidence level of $\approx 7\%$ for the more refined Roothaan wavefunction and only $\approx 0.3\%$ for the poorer M. B. S. wavefunction; a value of ≈ 0.1 results for the efficiency of each detecting channel.

Notwithstanding this low efficiency, ascribable to a misalignment of the apparatus and to the energy losses, this result confirms the potentiality of this technique in taking an insight into the real wavefunctions.

As to outer 2s and 2p electrons, which are not resolved in energy, from the angular distribution reported in Fig. 10 it is impossible to obtain a detailed information on e. m. d. function, due to the lack of experimental resolution.

It has to be pointed out that a misalignment of the two analyzers, which selects a non zero component of q along the direction perpendicular to the incident momentum p_0 , strongly affects the narrow momentum distribution of the L shell, peaked around the $q = 0$ value. The main effect is a lowering of counts, except for large q values. It has been computed that an out-of-plane of the two channels of 1 mm each and a total asymmetry of 1° in the angles reduces the counts on the maximum of the angular distribution, respect to a completely symmetrical condition, of a factor $K = 3$, in agreement with our experimental findings.

It has been also verified that such a misalignment, owing to the more flattened shape of the 1s Fourier transform, does not affect the measured angular distribution reported in Fig. 9.

In the mentioned conditions, the counts are lowered at half height of the L shell distribution (2° far from the maximum) by a factor $K = 2.7$. K reduces to unity only for the angles $31^\circ \geq \theta \geq 53^\circ$.

Accounting for these considerations, the characteristic width of the angular distribution for the external electrons can be inferred from the curve of Fig. 10. This results wider ($\leq 1^\circ$) than the value computed from a $2s2p^3$ Roothaan configuration, commonly employed in carbon bonding. Since this same broadening has previously been observed in Compton scattering⁽²⁰⁾ and in positron annihilation measurements, we confirm with this new technique that for external electrons in solid targets the momentum density is wider than expected for free atoms. Further work is in progress along this line.

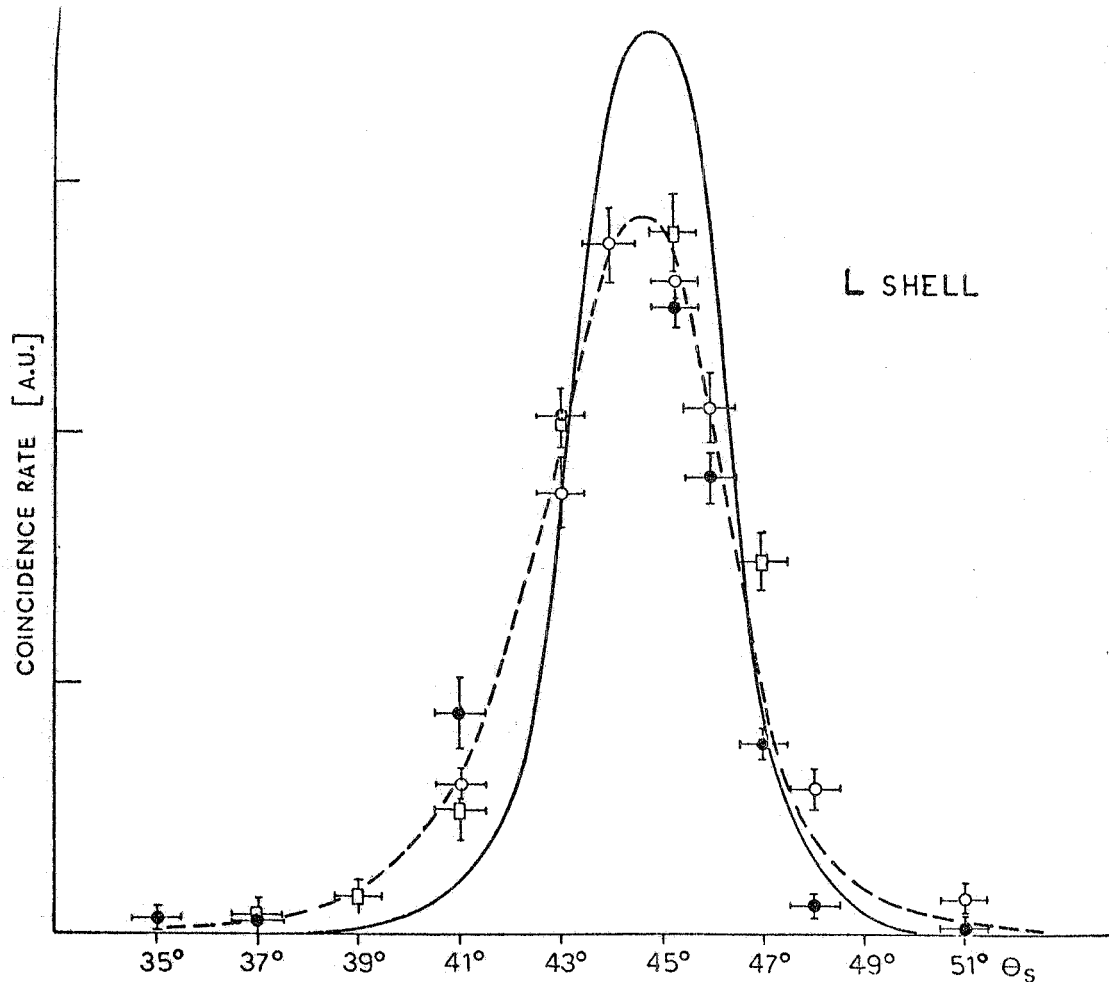


FIG. 10 - Angular distribution of the coincidence rate for the external L shell in Carbon films 250 (●) and 100 (○, □) Å thick. The data have been normalized to the same area. Experimental conditions are the same as in Fig. 9, but for the incident energy, which was $E_0 = 9.0$ KeV. The continuous curve is predicted in the basis of the Rothaan wave functions for external electrons, while the dotted curve is an eye interpolation to the measured points.

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