Measurement of the momentum distributions of *K* electrons of light atoms

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We report the results of an experiment on electron ejection by fast electrons in thin free-standing carbon and collodion films at primary electron beam energies of 10–11 keV. The energy spectrum reveals peaks due to ejection of *K* electrons of carbon, nitrogen, and oxygen, and also weakly bound outer electrons. For the *K* shells of carbon and oxygen we have measured the angular correlations of the ejected and scattered electrons, detected in coincidence. Analysis of the experimental data was carried out in terms of the impulse approximation, in which the behavior of the $(e,2e)$ cross section is determined by the shape of the momentum distribution of the electrons of a target with a given binding energy. The momentum distributions calculated by means of Hartree-Fock wave functions are in agreement with the measured angular correlations within experimental error.

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Measurement of the differential cross section for quasi-elastic knockout of an electron by a fast electron (e, 2e) is a new and promising method of studying the electronic structure of atoms, molecules, and crystals. As has been shown by Smirnov and Neudachin,[1,2] who first proposed the (e, 2e) method, complete kinematic information on the scattered and ejected electrons recorded in coincidence permits measurement of the momentum distributions of the electrons of the target. Here, in contrast to traditional methods (Compton profiles, positron annihilation, inelastic electron scattering at large angles), in which the integral distribution in momenta of all electrons of the target is studied, in the (e, 2e) method the binding energy of the ejected electron is measured simultaneously with its momentum. The nature of the electron momentum distribution in a given orbit is determined by the behavior of the corresponding single-electron wave function in the momentum representation. It is therefore possible to compare directly with experiment the various forms of wave functions proposed theoretically.

At the present time a number of experiments have already been carried out with the (e, 2e) method, both on solid-state[3] and gaseous[4,5] targets. In gas-target studies at comparatively low primary electron beam energies $E_0 \lesssim 1$ keV and a resolution $\Delta E \approx 2-4$ eV the momentum distributions and binding energies have been measured for electrons of the outer orbitals of a number of atoms[4] and molecules.[5] The high sensitivity of the method to the details of the behavior of the single-electron wave functions has been demonstrated, and also the possibility of studying finer effects associated with correlations of electrons in the target.

Experiments in solid-state targets require energies $E_0 \sim 10$ keV. This is due to the fact that at lower energies the interpretation of the experimental results is complicated as the result of multiple scattering effects even when the thinnest possible self-supporting films are used (~100 Å). At the same time the smallness of the cross section at these energies ($\sim 10^{-22}$ cm^2/eV-sr^2) does not permit use of small apertures for analysis of the electrons, which leads to a comparatively low experimental resolution. However, this is no obstacle in study of electrons of the inner shells with sufficiently high binding energy ϵ. In addition, for such problems solid-state films which provide a high density of particles are a more suitable target than a gas. Some groups[3] have used solid-state targets to study the momentum distribution of the K electrons of the carbon atom.

In this paper we present (e, 2e) experimental results in thin carbon and polymer films of thickness 200–300 Å for $E_0 = 10-11$ keV. We have used a symmetric experimental arrangement in which the momenta of the incident, scattered, and ejected electrons $\mathbf{p}_0$, $\mathbf{p}_1$, and $\mathbf{p}_2$ are coplanar, and electrons emitted at equal angles $\theta_1 = \theta_2 = \theta$ with respect to the primary beam direction and having identical energies $E_1 = E_2$ are detected in coincidence. In the case of ejection of K electrons of carbon and oxygen we have measured the angular correlations of the final electrons in the range of angles θ from 39 to 51°. The results obtained are compared with a calculation of the (e, 2e) cross section in the impulse approximation.

EXPERIMENTAL METHOD

A diagram of the apparatus is shown in Fig. 1. An electron beam was produced by a three-electrode long-focal-length directly heated electron gun 1, which was supplied with voltage from a highly stabilized power supply variable in steps of one volt in a working range of 10–15 kV. The beam current was varied over the range 10^{-12}–10^{-6} A. During the course of the measurements the current was monitored by means of a Faraday cup 2. The films studied were placed in 18 recesses in a special drum 3 located in the center of the chamber. The diameter of the region bombarded was 2.5 mm.

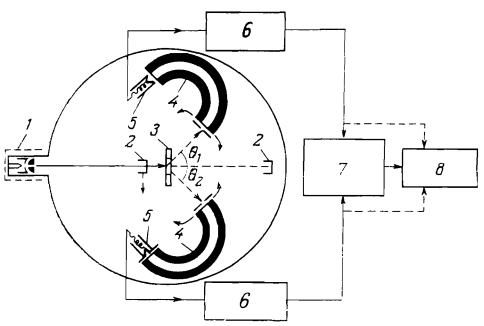

FIG. 1. Experimental arrangement.

Target changing was accomplished by rotation of the drum without breaking the vacuum. The chamber was pumped by a carbon sorption pump and then by magnetic ion pumps. The working pressure was 5×10^{-6} torr. Analysis and detection of the scattered and ejected electrons was carried out in two identical channels, each of which included a movable electrostatic energy analyzer 4, a detector 5, and an amplifier-shaper 6. The analyzers could be rotated around the center of the chamber in a range of angles from 0 to 60° relative to the primary beam direction.

The electrostatic analyzers consisted of hemispherical capacitors with R_{av} = 100 mm whose inner surfaces were blackened. The analyzing voltage was applied symmetrically with respect to ground potential. The detectors used were electron multipliers of the spiral channel type with a funnel at the input, which had a gain of $\sim 10^7$. To reduce the background produced by secondary electrons, the multipliers were supplied with voltage in such a way as to produce a retarding field U_{mult} = 3.6 kV which prevented arrival at the detector of electrons with lower energy. The intrinsic dark-current counting rate of a multiplier was 2—4 counts/sec. The signals at the multiplier anodes were fed to wide-band amplifiers with a gain K = 300, whose outputs were connected to shaping stages. Then the pulses were fed either to a coincidence circuit 7 and then to a scalar 8, or directly to the scalar in the case of measurements in one channel. The resolving time of the coincidence circuit was 20 nsec, which made it possible to overlap the asymmetry of the two detection channels, which was due mainly to the spread in the multiplier characteristics.

The targets were self-supporting films of carbon, collodion, and aluminum of thickness 180—350 Å. The carbon and aluminum films were obtained by evaporation onto a substrate which was then dissolved in water. The polymerized collodion films were prepared from a solution of collodion in amyl acetate. The film thickness was measured in a UM-2 monochromator with a special wedge attachment.[6] As a result of the fact that the method used did not guarantee either uniformity of the target in thickness or the absence of microcracks, the suitability of each sample was checked in practice by measurements in one channel of electrons scattered by a given target at an angle $\theta \sim 45°$. In each such spectrum a narrow intense peak due to elastic scattering of electrons by nuclei was observed at $E_1 \approx E_0$, and also a broader peak with a maximum at $E_1 \approx E_0 \cos^2\theta$ corresponding to quasifree scattering by electrons in the target. A film was considered suitable for further measurements if the electron peak was at least twice background.

To clarify the role of multiple collisions we used the results of experiments on transmission through aluminum films of different thicknesses. The distributions obtained for $E_0 \approx 10$ keV are shown in Fig. 2. It follows from these results that on passage through films of thickness less than 340 Å the intensity of the primary beam drops by several times but the spreading of the beam in energy and angle are no greater than 20 eV and 9×10^{-4} rad, respectively. We can expect that these results are valid in order of magnitude also for the polymerized and carbon films. In the coincidence measurements the energy resolution was 140 eV and the angular resolution 3×10^{-2} rad. Therefore we did not take into account these spreads in interpretation of the (e, 2e) experimental results.

EXPERIMENTAL RESULTS AND DISCUSSION

In the range of angles $39° \leq \theta \leq 51°$ we measured the dependence of the coincidence counting rate on the primary beam energy E_0 for fixed values of final electron energy. Figure 3 shows typical results obtained in carbon films of thickness 250 Å at θ = 45 and 51°, $E_1 = E_2$ = 5025 eV, and a primary beam current $I_0 = 10^{-7}$ A. From the conservation of energy it follows for the process considered $E_0 = E_1 + E_2 + \epsilon$ that the maximum at E_0 = 10 330 eV corresponds to ejection of K-shell electrons of carbon with a binding energy ϵ = 280 eV. As a result of the inadequate resolution, the single combined peak at $E_0 \approx$ 10 050 eV = $E_1 + E_2$ corresponds to ejection of the remaining electrons, which have significantly lower binding energies. For the K electrons of carbon, Fig. 4 shows the dependence of

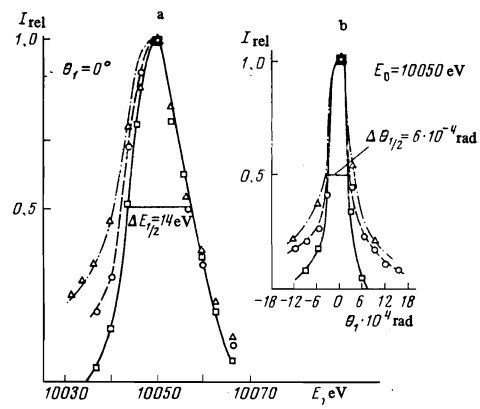

FIG. 2. Energy distribution (a) and angular distribution (b) of electrons which have passed through aluminum films: Δ) target thickness 340 Å ($I_{max} = 1.2 \times 10^{-9}$ A); ○) target thickness 180 Å ($I_{max} = 4.3 \times 10^{-9}$ A); □) primary beam ($I_{max} = 5.5 \times 10^{-9}$ A). $I_{rel} = I_{det}/I_{max}$.

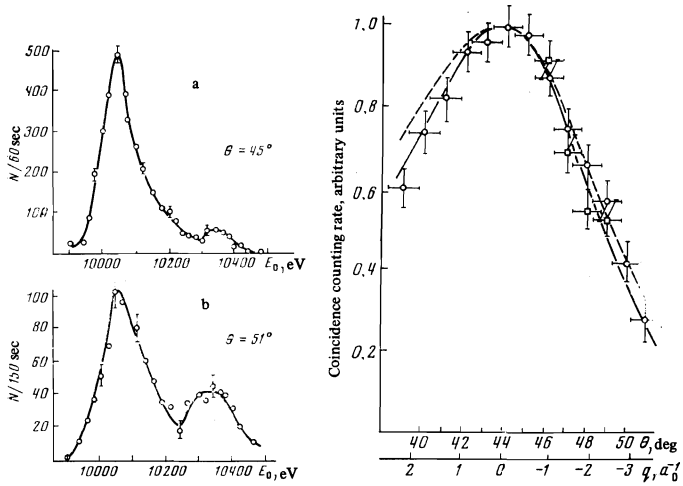

FIG. 3. Dependence of coincidence counting rate on primary beam energy, obtained in a carbon film of thickness 250 Å for $E_1 = E_2$ = 5025 eV, $I_0 = 10^{-7}$ A: a) θ = 45°, b) θ = 51°. N is the number of coincidences.

FIG. 4. Angular correlations of final electrons for K shell of carbon: ○) measurements in carbon films, □) measurements in collodion targets, solid line—calculation with Hartree-Fock wave function, [7] dashed line—calculation with Slater type function, Z_{1S}^C = 5.67.

the coincidence counting rate on the emission angle of the final electrons. Since the intrinsic width of the hole levels of the K electrons is significantly less than the energy resolution of the apparatus, the various points in Fig. 4 were obtained by calculation of the areas of the corresponding peaks in the energy dependence of the coincidence counting rate, measured at various values of θ.

In Fig. 5 we have shown the coincidence energy spectrum obtained in a collodion film of thickness 250 Å at $\theta = 48°$, $E_1 = E_2 = 5150$ eV, and $I_0 = 2 \times 10^{-8}$ Å. The polymerized collodion film is an incomplete cellulose ether $[C_6H_7O_2(ONO_2)_3]_n$ in which 20–30% of the molecules have an (OH) group instead of an (ONO_2) group. Therefore it is clear that the peaks observed in the coincidence spectrum at $E_0 = 10\,580$, 10 700, and 10 830 eV are due respectively to ejection of K electrons of atoms of carbon ($\epsilon = 280$ eV), nitrogen ($\epsilon = 400$ eV), and oxygen ($\epsilon = 530$ eV). The single common peak at $E_0 \approx E_1 + E_2$ corresponds to ejection of the remaining electrons with smaller binding energies.

Unfortunately, as a result of the relatively small percentage content of nitrogen in the target, the K level of nitrogen did not appear very clearly. The large uncertainty in the area of the corresponding peaks (at $\theta = 48°$ in Fig. 5 we can see a flattening, and at smaller angles this peak appears even more weakly) did not allow us to obtain the angular dependence of the coincidence counting rate for ejection of nitrogen K electrons. The corresponding angular dependence for ejection of oxygen K electrons is shown in Fig. 6. Some results for K electrons of carbon, which enters into the composition of the collodion target, are shown in Fig. 4, where they can be compared with the data obtained in carbon films. We note that they agree within the experimental error.

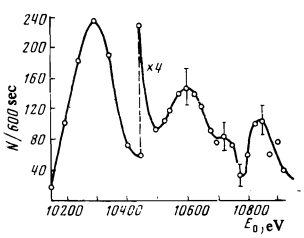

FIG. 5. Dependence of coincidence counting rate on E_0, obtained in a collodion film of thickness 250 Å, for $\theta = 48°$, $E_1 = E_2 = 5150$ eV, $I_0 = 2 \times 10^{-8}$ A.

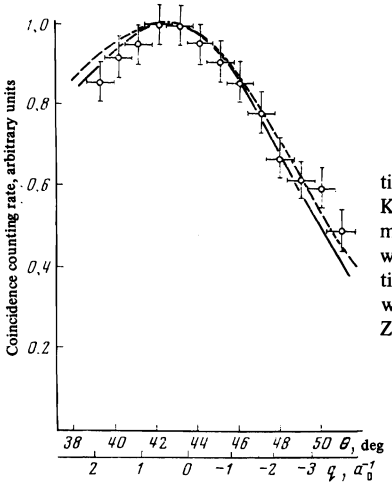

FIG. 6. Angular correlations of final electrons for the K shell of oxygen: ○) experiment, solid line) calculation with Hartree-Fock wave function, dashed line) calculation with Slater type wave function, $Z^0_{1S} = 7.66$.

The experimental data can be analyzed in terms of the impulse approximation,[2,3] which is applicable under our conditions, since even the strongly bound electrons of the targets used satisfy the relations

$$\varepsilon \ll E_0, \quad \langle q \rangle \ll |\mathbf{p}_0 - \mathbf{p}_1| = |\mathbf{p}_0 - \mathbf{p}_2|, \tag{1}$$

where $\langle \mathbf{q} \rangle$ is the average momentum of electrons with a given binding energy ϵ. In this approximation the cross section for the (e, 2e) reaction is written in the form[2]

$$\frac{d\sigma}{d\Omega_1\, d\Omega_2\, dE_1} = \frac{mp_2}{\hbar^3}\left(\frac{d\sigma}{d\Omega_1}\right)_{\text{lab}} |\overline{F(q)}|^2, \tag{2}$$

where $\hbar\mathbf{q} = \mathbf{p}_1 + \mathbf{p}_2 - \mathbf{p}_0$; $(d\sigma/d\Omega)_{\text{lab}}$ is the cross section for free scattering of an electron by an electron in the laboratory system; $F(q)$ is a form factor, which in the self-consistent-field approximation for the many-electron wave functions of the atom and the residual ion reduces to the Fourier transform of the single-particle wave function of the ejected electron

$$\varphi_{nlm}(r) = R_{nl}(r)\, Y_{lm}(\hat{r}),$$
$$F_{nlm}(\mathbf{q}) = \frac{1}{(2\pi)^{3/2}} \int e^{-i\mathbf{q}\mathbf{r}} \varphi_{nlm}(\mathbf{r})\, d\mathbf{r} = \varphi_{nlm}(\mathbf{q}). \tag{3}$$

Here the quantity $|\overline{F(q)}|^2$ takes the form

$$|\overline{F_{nl}(q)}|^2 = \frac{1}{2l+1} \sum_{m=-l}^{l} |\varphi_{nlm}(\mathbf{q})|^2 = \frac{2}{\pi} \left| \int j_l(qr) R_{nl}(r) r^2\, dr \right|^2, \tag{4}$$

where $j_l(x)$ is the spherical Bessel function. In this way, by measuring the cross section (2), we obtain the distribution of electrons in momentum in an orbit with a given binding energy $\epsilon_{nl} = E_0 - E_1 - E_2$. In the symmetric variant of the experiment which was used, the contribution to the cross section is only from target electrons whose momenta are parallel to the primary beam. From the conservation laws we have for the value of this momentum

$$\hbar q = \sqrt{2mE_1}\,(2\cos\theta - \sqrt{2 + \varepsilon/E_1}). \tag{5}$$

The results of calculation of the angular correlations of the final electrons with Eqs. (2) and (4) are shown in Figs. 4 and 6. For the wave functions of the K electrons of carbon and oxygen we used the functions of the Hartree-Fock approximation[7] (solid curves) and the simpler functions of the Slater type with effective charges $Z^C_{1S} = 5.67$ and $Z^O_{1S} = 7.66$ (dashed curves). For convenience in comparing the theoretical and experimental data, we have shown on the abscissa of the plots scales of angles and momenta, which are related by expression (5). We note that the asymmetry of the angular correlation curves with respect to the value $q = 0$ (the corresponding angle is $\theta_0 = \arccos \sqrt{(1 + \epsilon/2E_1)/2}$) is due to the nature of the dependence of the cross section $(d\sigma/d\Omega)_{\text{lab}}$ which enters into Eq. (2), on the scattering angle θ.

From the figures it is evident that the calculation with the Hartree-Fock functions agrees with the measurements within the experimental error. At the same time, the use of the Slater functions leads to some disagreement of the theoretical and experimental results, which is appreciable for $\theta \leq 41°$. This indicates that even with comparatively poor energy resolution the (e,2e) method permits us to judge the quality of the various wave functions used for description of the single-electron states.

Substantial interest is presented by measurements

of the momentum distributions of the electrons located in an outer energy level, and also of the free electrons in the crystal.[8] In order to be able to carry out such experiments, work is going on at the present time to increase the energy resolution of the apparatus without significant loss in aperture.

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