

- Phys. Dokl. 19, 288 (1974)].
- ¹⁷E. M. Dianov, A. Ya. Karasik, V. B. Neustruev, A. M. Prokhorov, and I. A. Shcherbakov, Dokl. Akad. Nauk SSSR 224, 64 (1975) [Sov. Phys. Dokl. 20, 622 (1975)].
- ¹⁸A. I. Burshtein and A. Yu. Pusek, Fiz. Tverd. Tela (Leningrad) 16, 2318 (1974) [Sov. Phys. Solid State 16, 1509 (1975)].
- ¹⁹T. T. Basiev, Yu. K. Voron'ko, and I. A. Shcherbakov, Zh. Eksp. Teor. Fiz. 66, 2118 (1974) [Sov. Phys. JETP 39, 1042 (1974)].
- ²⁰T. T. Basiev, Yu. K. Voron'ko, T. G. Mamedov, and I. A. Shcherbakov, Kvantovaya Elektron. (Moscow) 2, 2172 (1975) [Sov. J. Quantum Electron. 5, 1182 (1975)].
- ²¹B. F. Ormont, *Struktura neorganicheskikh veshchestv* (Structure of Inorganic Substances), Gostekhizdat, 1950.
- ²²T. Miyakawa and D. L. Dexter, Phys. Rev. B1, 2961 (1970).
- ²³Yu. K. Voron'ko, V. V. Osiko, and I. A. Shcherbakov, Zh. Eksp. Teor. Fiz. 63, 691 (1972) [Sov. Phys. JETP 36, 365 (1973)].
- ²⁴D. L. Dexter, J. Chem. Phys. 21, 836 (1953); A. S. Agabekyan, Proc. 3rd All-Union Seminar on Nonradiative Energy Transfer in Condensed Media, Erevan, 1950, p. 53.
- ²⁵D. W. Posener, Aust. J. Phys. 12, 184 (1959).

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Scattering of electrons by inert gas atoms with ejection of one or two electrons from the outer shell

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Beam electrons inelastically scattered by He, Ne or Ar atoms are separated by a coincidence technique into components due to single or double ionization of the outer shells. The initial electron energy is 4 keV and the scattering angle range is 0-10°. The energies transferred do not exceed the ionization threshold for the inner shells. The absolute values of the differential cross sections for single and double ionization are measured as functions of two variables, the transferred energy and the scattering angle. The generalized oscillator strengths (GOS) for single and double ionization are determined from the measured momentum transfers up to 3 atomic units. At small values of the transferred momenta the GOS are consistent with available optical data for both single and double ionization. The dependences of the double-ionization GOS on the transferred momentum has a characteristic peak whose position is practically identical with that of the single-ionization GOS. On the other hand a difference in the shapes of the curves of the single and double ionization GOS as a function of momentum transfer is observed. The magnitude and character of the difference depend on the atom. The narrower GOS peak is related to double ionization via the virtual Auger effect. The argon single-ionization GOS are calculated in the RPAE approximation. It is shown that the calculation, in effect, also takes into account multiple ionization.

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1. INTRODUCTION

One of the methods of investigating the structures of atoms and molecules is to study the angular and energy distributions of the electrons scattered by them.^[1,2] At sufficiently high electron energies, these experiments yield the generalized oscillator strengths (GOS), which are connected in the Born approximation with the differential inelastic-scattering cross sections.^[3] The GOS depend on two parameters, the energy and momentum given up in the collision, and the set of these parameters constitutes a two-dimensional spectrum that provides a more complete description of the target^[4,5] than the optical photoabsorption spectrum; the latter is the limiting case of the GOS at zero momentum transfer. The GOS measured in this manner turn out to be summed over the final states of the target.

The influence of the electron-electron interactions in the atom (correlations) on the elastic scattering of fast electrons was explained in earlier papers.^[1,4,5] In the present paper we study the inelastic-scattering components that are connected with a definite multiplicity of

the target ionization and have been separated by the coincidence method.

Multiple ionization, as all many-electron transitions in general, is due to the interaction of the target electrons with one another. Even to explain the existence of such transitions it is necessary to take the interelectron interactions into account in greater detail than in the Hartree-Fock method (assuming the self-consistent field to be the same).^[6] The oscillator strengths of many-electron transitions have been investigated up to now, both theoretically and experimentally, only in the optical limit.

Attempts at a quantitative description of two-electron transitions are being undertaken in two directions—by taking into account the interelectron correlations in the wave functions, as well as with the aid of many-body quantum theory. The first of these methods is limited to light atoms (results are available for He^[7,8]) and its success depends on the proper choice of the form of the wave functions. The second method^[9] is more consistent and is suitable for both light^[10] and heavy^[9,11]

atoms. The number of such calculations, however, is small, as are also the experiments with which the calculations could be compared.

The experimental investigations of many-electron transitions to the continuum, due to the incident electrons, were confined up to 1971 to measurements of the integral multiple-ionization cross sections^[12] or the cross sections for ionization with excitation.^[13] Subsequently, Van der Wiel and co-workers^[14] investigated by a coincidence procedure the cross section for single and multiple ionization of inert gases in the scattering of 10-keV electrons through zero angle as a function of the energy transfer. However, the optical oscillator strengths calculated by Van der Wiel *et al.*^[14] from the results of these measurements within the framework of the Born approximation differ noticeably (by up to a factor of two) from the values directly measured in experiments with photons.^[15,16] It remained unclear whether this difference was due to the experimental error or whether it was fundamental in character.

The present paper deals with removal of one and two electrons from the outer shell of the target at momentum transfers up to 3 at. un. The first investigations in this direction were already reported by us in^[17]. We registered coincidences of scattered electrons with the produced ions. Multiple ionization due to the Auger effect was excluded because the chosen values of the energy transfers were insufficient for the ionization of the internal shells. The doubly-differential (with respect to angle and energy transfer) cross sections for the scattering of 4-keV electrons with single and double ionization of He, Ne, and Ar were measured in absolute units. The experiments were performed in the scattering-angle range from 0 to 10°. In the limiting case of small angles, the results are compared with the available values of the optical oscillator strengths.

2. EXPERIMENT

Experimental setup. The measurements were performed with a setup adapted both for the study of inelastic scattering of electrons at a fixed charge of the produced ion, and for absolute measurements of the total yield of ions with different charges. The experimental setup is illustrated in Fig. 1. An electron beam from source So is collimated by a series of slits S₁-S₄, enters the collision chamber C filled with the investigated gas and leaves this chamber together with the scattered electrons through a broad slit S₅. The primary current is measured with a movable Faraday receiver F₁, which is removed when the scattered electrons are registered. A parallel-plate capacitor is placed in the collision chamber. One of its plates has a rectangular slit 2×20 mm cut along the axis of the electron beam. The second electrode of the capacitor is a grid with a transmission 0.94, through which the gas flows into the ionization region. The slow ions produced in the collision chamber are drawn out through the slit by the electric field of the capacitor and are then accelerated by a uniform electric field produced by an electrode system Dr. The accelerated ions are focused with an immersion lens onto the entrance slit

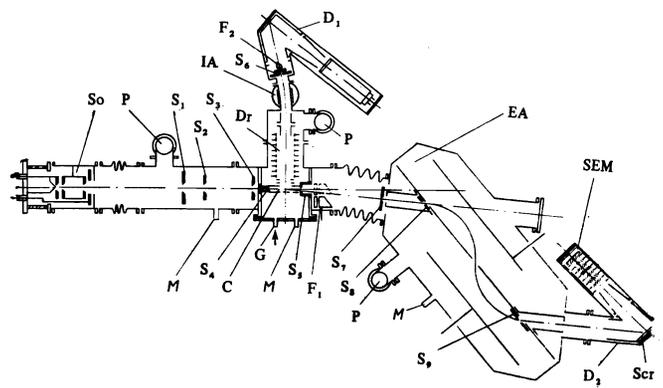


FIG. 1. Diagram of experimental setup.

S₈ of the ion detector. Between the lens and the ion detector is a magnetic analyzer IA, which separates the ions of the required charge. The ion detector consists of a Faraday receiver with a lifting bottom and a particle counter^[18] D₁ located behind the Faraday receiver. This detector makes it possible to measure the number of ions both in the current-flow regime and in the regime wherein individual particles are counted. The ion loss over the entire path does not exceed 6%. To decrease the effect of the stray magnetic field on the electron beam, an iron-clad magnet with compensating coils was used. The scattered electrons were analyzed by a rotating part of the installation, containing the scattered-electron collimator S₇-S₈, an electrostatic energy analyzer EA, and electron counter D₂. The relative resolution of the analyzer EA was 10⁻³. The resultant angular resolution of the installation was not worse than 0.7°. The working concentration of the target atoms in the collision chamber was maintained by continuously feeding the gas to pipe connection G and drawing it with three pumps P.

The pulses from the particle counters were fed to a delayed-coincidence circuit. A feature of the experiment was the need for the separating of the true coincidences against the background of a large number of random coincidences that occur when the scattered electrons due to double ionization are registered. To take this background into account, we used a coincidence circuit that delivered continuously to an additional output, during the course of the entire operating time, a sequence of pulses simulating random coincidences. The principle underlying such a system was described by Petushkov and Melioranskiĭ.^[19] The pulses from the outputs of the coincidence circuit and from each of the detectors were registered by scaling instruments which were connected with an automatic printout device.^[20] The printout device received also data from a control block that varied the energy-analyzer voltage in accordance with a prescribed program.

Measurement procedure. The number of ions n_i , the number of scattered electrons n_e , and the number of two coincidences q registered per unit time are proportional to the corresponding cross sections and are determined by the relations

$$n_i = \frac{I}{e} N l_i \alpha_i \sigma^{k+}, \quad (1a)$$

$$n_e(E, \theta) = \frac{I}{e} N l_e \omega_e \alpha_e \frac{\partial^2 \sigma}{\partial E \partial \Omega}(E, \theta), \quad (1b)$$

$$q(E, \theta) = \frac{I}{e} N l_i \omega_i \alpha_i \alpha_e \frac{\partial^2 \sigma^{k+}}{\partial E \partial \Omega}(E, \theta). \quad (1c)$$

Here I/e is the flux of primary electrons, N is the concentration of the target atoms in the collision chamber, l_i and l_e are respectively the lengths of the sections in which the ions and scattered electrons were gathered, ω_i and ω_e are the solid angles, averaged over these sections, in which the scattered electrons and ions were gathered, α_i and α_e are the efficiencies of the gathering and registration of the ions and electrons, σ^{k+} are the integrated k -fold ionization cross section, $\partial^2 \sigma / \partial E \partial \Omega$ is the differential cross section for inelastic scattering of an electron through an angle ϑ with a transfer of energy E to the target, and $\partial^2 \sigma^{k+} / \partial E \partial \Omega$ is the differential cross section for scattering with k -fold ionization of the target.

The absolute values of the integrated cross sections σ^{k+} were determined from relation (1a). These measurements were described by us earlier.^[21] The differential cross sections with k -fold ionization, as can be easily seen from (1), can be obtained from the formula

$$\frac{\partial^2 \sigma^{k+}}{\partial E \partial \Omega} = \sigma^{k+} \frac{q}{n_i} \frac{1}{\omega_i \alpha_e}. \quad (2)$$

The average electron-gathering solid angles ω_i and ω_e , as shown by analysis of the geometry of the experiment, could be regarded as equal to each other and constant in our range of scattering angles.

The experimental geometry was verified with the aid of control experiments that are analogous in principle to those described by Afrosimov *et al.*^[22] The pressures did not exceed $(2-15) \times 10^{-4}$ mm Hg in the collision chamber and 5×10^{-6} mm Hg outside the chamber. In the measurements with helium we used the isotope ^3He , thereby facilitating the registration of the He^{2+} ions against the apparatus background of the H_2^+ ions. To reduce the experiment time, the cross section for electron scattering through angles larger than 3° with single ionizations of the target were calculated as the difference between the cross section for the total scattering (without determining the final state of the target) and the cross sections for scattering with double ionization of the target. In the measurements of the total-scattering cross sections we used the calculated dependence of the length of the gathering region of the scattered electrons on the scattering angle. The validity of this procedure was verified for one of the gases (neon), for which the scattering cross sections were measured in the entire range of scattering angles in coincidence with both singly-charged and doubly-charged ions.

Experimental errors. The errors in the measurement of the doubly differential cross sections are determined by the measurement errors and by the drifts of the parameters that enter in relation (2). The errors in the previously measured integrated cross sections

σ^{k+} were estimated in^[21]. Since $n_i \gg q$, the error in the measurement of the ratio q/n_i was determined by the statistical accuracy of the measurement of q , which, together with the error in the measurement of σ^{k+} , determines in fact the errors of the entire experiment. The instability of the electron counting efficiency did not exceed 10%. At small scattering angles, a noticeable source of errors is the uncertainty in the determination of this angle, due to the uneven density of the primary beam in the collimator, to the influence of the surface charge at the slits, etc. This uncertainty is estimated by us at 0.1° . The contribution of the foregoing factors was taken into account in the calculation of the estimated errors cited in Sec. 3.

Absolute calibration. The relative values of the doubly differential single- and double-ionization cross sections obtained with the aid of relation (2) were converted into absolute units by normalizing the associated generalized oscillator strengths (see relation (4)) in accordance with a sum rule wherein for each fixed value of the momentum transfer K there is satisfied the relation^[3]

$$\int_0^\infty \frac{\partial f}{\partial E}(K, E) dE = z, \quad (3)$$

where z is the number of electrons in the atom. Approximately similar relations are satisfied for each shell of the atom separately. The most convenient for the normalization is the GOS of neon, since on the one hand, their values are not too small, and on the other, eight of the ten electrons of the neon atoms belong to its outer shell; the two internal electrons have greatly different binding energies and the approximate sum rule for the outer shell can therefore be expected to be satisfied in this case with the greatest accuracy.

The absolute calibration of the installation was carried out in accordance with the approximate sum rule for the external shell of neon. We integrated the GOS corresponding to scattering through angles 5.7 and 0.45° . In the former case, as seen from relation (4), the momentum transfer depends little on the energy transfer E and can be regarded as constant for the entire region E in which the measurements were carried out. In the second case this quantity is close to its minimum (for a given E) value and varies strongly with changing E . In accordance with the general behavior of the GOS of the continuous spectrum,^[3] we can expect its dependence on the momentum transfer to be insignificant under these conditions. Substitution in (3) of the GOS corresponding to a constant small scattering angle, instead of a constant momentum transfer, is equivalent to neglecting this dependence. The results of the calibrations carried out in these two cases agree within the limits of the experiment error. In the integration up to $E = 400$ eV we used the GOS obtained from the measurements; for larger E we used an extrapolation in accordance with the $E^{-7/2}$ law. In both cases the contribution of the extrapolated part to the total GOS sum did not exceed 5%. The total absolute-calibration error is estimated by us at not more than 7%.

3. RESULTS AND DISCUSSION

Differential cross sections. The measured doubly differential cross sections $\partial^2\sigma^{k+}/\partial E\partial\Omega$ for the scattering of electrons with single and double ionization of He, Ne, and Ar are shown in Figs. 2 and 3. Experiments in which all these cross sections would be taken into account in a sufficiently wide range of scattering angles¹⁾ were not performed previously, and our results are new. The dominant inelastic process in the investigated range of conditions is single ionization. The relative contribution of double ionization increases on going to heavier atoms. With Ne as an example (Fig. 2), we see that the character of the variation of the yield of the doubly-charged ions remains the same with increasing energy transfer on going from small (0.45) to appreciable (5.7°) scattering angles. In both cases the cross section, after reaching a maximum at a value E larger by 1.5–2 times than the double ionization threshold, begins to decrease and remains proportional to the corresponding single-ionization cross section. At a fixed value of E , the order of magnitude of the relative yield of the doubly charged ions remains constant in the investigated range of scattering angles.

At scattering angles larger than 3° (Fig. 3) there are certain differences between the forms of the angular distributions of the single and double ionization cross sections. The magnitude and character of these differences vary from atom to atom. For neon, the existence of the observed difference was predicted by us in^[17] on the basis of the result of an experiment in which we investigated the angular distribution of the electrons scattered in single and double ionizations, without an analysis of the energy transfer. In the same reference we noted the connection between this difference and the effective mechanisms of double ionization.

It is more convenient to continue the discussion of the results and their comparison with other available data in terms of the GOS.

Generalized oscillator strengths. The GOS are connected with the measured cross sections, assuming the Born approximation to be valid, by the relation^[3]

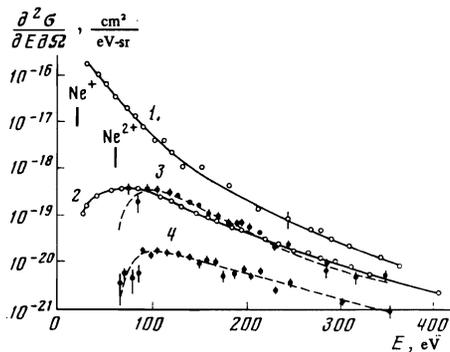


FIG. 2. Energy-loss spectra in electron scattering by Ne through fixed angles with single and double ionizations of the target. $E_0 = 4$ keV. 1) (light circles)— Ne^+ , 0.45°; 2) (light circles)— Ne^+ , 5.7°; 3) (dark circles)— Ne^{2+} , 0.5°; 4) (dark circles)— Ne^{2+} , 5.7°. Fiducial probabilities 70%.

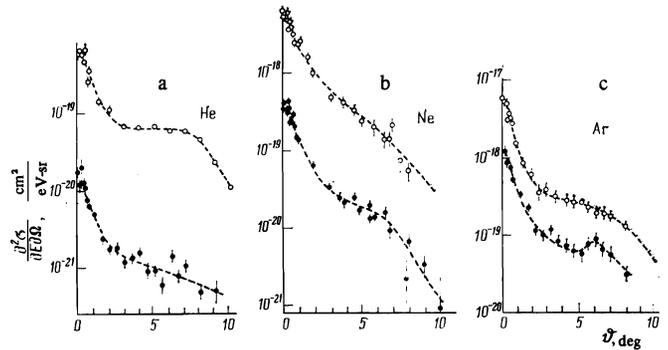


FIG. 3. Angular dependences of the doubly differential cross sections for the scattering of electrons with single and double ionization of the target at fixed energy transfer E . a) He, $E = 115$ eV; b) Ne, $E = 115$ eV; c) Ar, $E = 85$ eV. Light circles—single ionization, dark circles—double ionization. $E_0 = 4$ keV. Fiducial probabilities 70%.

$$\frac{\partial f}{\partial E}(E, K) = \frac{EK^2 k_0}{2 k'} \frac{\partial^2 \sigma}{\partial E \partial \Omega}(E, \theta), \quad (4)$$

where

$$K^2 = 4(2E_0 - E) \sin^2 \frac{\theta}{2} + \frac{E^2}{2E_0} \cos \theta,$$

$\partial f/\partial E$ is the spectral density of the generalized oscillator strength, E and K are the energy and momentum transferred in the collision, E_0 is the energy of the incident electrons, k_0 and k' are the momenta of the incident and scattered electrons, and θ is the scattering angle. The atomic system of units is used.

At present there are no quantitative criteria whatever for the applicability of the Born approximation to the description of the inelastic-scattering component connected with the double ionization. Let us examine the possibility of determining the double-ionization GOS with the aid of relation (4) under our conditions. The processes that occur in the scattering are graphically illustrated in Fig. 4. Graph a describes single ionization, the dominant contribution of which can determine the possibility of successfully describing the total scattering in the Born approximation. For double ionization there are mechanisms that fit in the framework of the Born approximation (diagrams c–f), as well as mechanisms beyond its limits (diagram b). In the former case, double ionization is due to the interaction of the target electrons with one another, and in the latter they are due to the double interaction of the incident electron with the target electrons. With increasing initial energy, the cross section of process b decreases like E_0^{-2} , whereas the Born cross sections (graphs c–f) decrease like E_0^{-1} . Consequently, if E_0 is large enough, the contribution of graph b can be neglected. We shall calculate the double ionization GOS, just as that of single ionization, with the aid of relation (4), assuming the energy E_0 to be large enough to use the Born approximation. The validity of this assumption will be checked later on against the agreement between the results and the values of the oscillator strengths measured in experiments with photons.

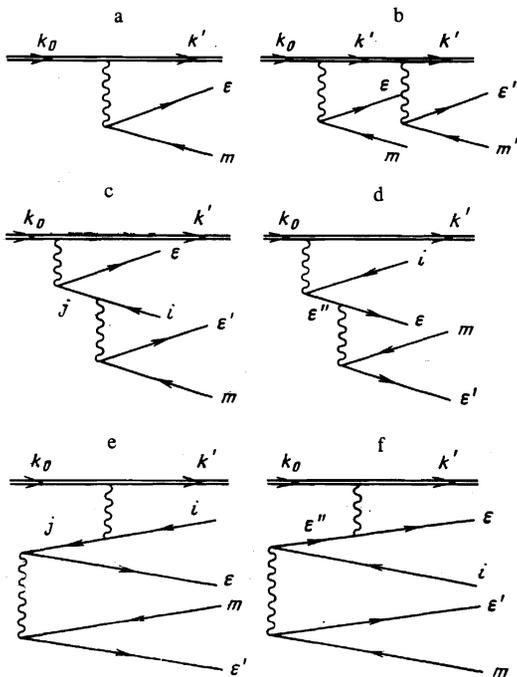


FIG. 4. Graphs of the processes in inelastic scattering of electrons by an atom. A double line denotes a fast electron described by a plane wave, while simple lines denote the hole (vacancy) produced in the atom or the knocked out (slow) electron, while the wavy line represents the Coulomb interaction. The double-ionization processes are shown in the lowest order of perturbation theory in the electron-electron interaction.

Limiting values of GOS. The GOS values obtained from the measured cross sections with the aid of relation (4) and extrapolated to $K=0$, together with results of other studies, are summarized in Table I. It can be seen that the limiting single-ionization GOS obtained by us agree reliably, within the estimated errors, with the optical oscillator strengths obtained from experiments both with photons and with electrons. As to double ionization (and the ratio $(df^{2+}/dE):(df^+/dE)$), we see here a great discrepancy between results of different experiments. Our results agree satisfactorily with the results of experiments with photons. The deviation from Van der Wiel's results is connected, in our opinion, with the errors of his experiment.²⁾

Let us compare our values of the double-ionization GOS with the theoretical calculations of the double-ionization oscillator strengths available for He and Ne. Figure 5 shows the GOS spectrum obtained from the spectrum of the energy lost by electrons scattered from Ne through an angle 0.45° and registered by coincidence with doubly charged ions. As seen from the figure, there is qualitative agreement with the calculation of Chang and Poe,^[11] who used many-body quantum-theory methods. As seen from the table, the values for df^{2+}/dE of helium agree with a calculation^[8] in which the correlations were taken into account in the wave function of the ground state and the matrix elements of the velocity operators were used. However, the large discrepancy between the results of this calculation, obtained with investigation of the matrix elements of the

TABLE I. Optical oscillator strengths of single and double ionization.

Atom	E, eV	$\sum \frac{df^{k+}}{dE}, 10^{-3} eV^{-1}$	$\frac{df^+}{dE}, 10^{-3} eV^{-1}$	$\frac{df^{2+}}{dE}, 10^{-3} eV^{-1}$	$\frac{df^{2+}/dE}{df^+/dE}, \%$	Method	Reference
He	115	2.80 ± 0.77	2.75 ± 0.75	6.1 ± 1.9	2.2 ± 0.8	Experiment (electron)	Present work [14]
			2.78	13.2	4.7	" "	" "
					3.6*	" "	" "
					2.14	" "	" "
Ne	115	26.8 ± 1.5	25 ± 1.5	175 ± 29	7.0 ± 1.2	Experiment (electrons)	Present work [14]
			23.8	261	11	" "	" "
					8.5*	Experiment (photons)	[15]
					8.7	" "	" "
Ar	85	12.5 ± 2.0	10.2 ± 1.9	230 ± 50	23 ± 6	Experiment (electrons)	Present work [14]
			9.3	309	33	" "	" "
					20*	Experiment (photons)	[15]
					14	" "	" "
			21	" "	" "		
		13.8 ± 0.5				RPAA calculation	Present work [28]
		13.2					Present work [8]

Notes. 1. The indicated errors of the quantities measured by us correspond to a fiducial probability 90%. 2. Values marked by an asterisk were obtained from Van der Wiel's results (experiment with electrons), which were kindly supplied by the author.

velocity and the length operators (the calculations with ∇ and r) indicate that this calculation is not reliable.

Dependence of GOS on the momentum transfer. The experimentally obtained single and double-ionization GOS of He, Ne, and Ar as functions of the momentum transfer at a fixed energy transfer are shown in Fig. 6. We consider first single ionization. In the case of He, owing to the relative smallness of the double-ionization cross section, the single-ionization GOS practically coincide with the inelastic-scattering GOS. The latter were measured in experiment on inelastic scattering of electrons^[2] with $E_0 = 25$ keV without recording the final state of the target. Our results agree within 15% with the data of^[2]. The GOS curves for all targets have a pronounced maximum. This maximum is a characteristic feature of the single-ionization GOS^[3] and is due to the transfer of the entire energy to one electron re-

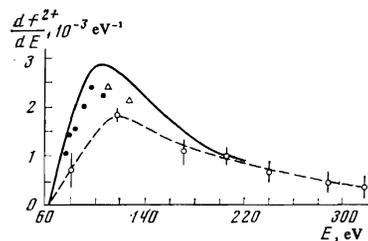


FIG. 5. Oscillator strengths for double ionization of neon. Light circles and solid line—obtained from scattering of electrons with $E_0 = 4$ keV through 0.5° . The vertical bars denote the relative measurement errors. The error of the absolute calibration is 13%. The fiducial probabilities are 90%. Dark circles—obtained optical data^[16] and^[27], triangles—from optical data of^[15] and^[27].

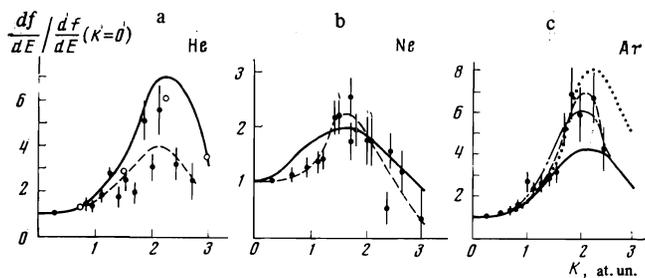


FIG. 6. Dependence of the generalized oscillator strengths of single and double ionization on the momentum transfer K at a fixed energy transfer E : a) He, $E = 115$ eV; b) Ne, $E = 115$ eV; c) Ar, $E = 85$ eV. Solid curves—single ionization, dashed curves and dark circles—double ionization. Light circles—combined scattering (from the experiment of [21], dash-dot curve—removal of any electrons from outer shell of Ar (RPAE calculation), dashed curve—removal of $3s$ electron (RPAE calculation).

moved from the target. The width of the maximum is determined by the momentum representation of the atomic electron in the bound state.

In the case of Ar, for comparison with the experimental results, we calculated the single-ionization GOS in the random-phase approximation with exchange (RPAE). The calculation was performed by the method described in detail in [4, 5, 23] in accordance with the programs of [24, 25]. Account was taken of the contribution of the transitions in which the change Δl of the orbital angular momentum was equal to 0, 1, 2, 3, 4, and 5. Just as before, [5] in this calculation account was taken of the correlations between the electrons of one subshell and, in addition, between electrons of different subshells ($3s$ and $3p$), as well as different subshells (the influence of the $2s$ and $2p$ electrons on $3s$ and $3p$). As shown in an earlier study of photoionization, [23] the correlations between the subshells and between the shells may exert a strong influence on the oscillator strengths. The most significant is the influence of the correlation on the dipole ($\Delta l = 1$) and monopole ($\Delta l = 0$) transitions. The correlations between shells at the given $E = 85$ eV turn out to be small. Figure 7 shows the GOS curves for single-ionization and for the sum of the inelastic scattering of electrons, obtained from our experiments, together with the results of the described calculation. It is seen that out of the two experimental curves, the summary-scattering GOS curve agrees better with the calculation. To understand these facts one must bear it in mind that the RPAE method does not take into account at all the existence of more than one particle-hole pair in the final state. In the RPAE, at the same time, a sum rule is satisfied, and in the case of the exact description it holds for the sum of the GOS of the transitions to all the final states. This makes a discrepancy between the calculation and the experimental single-ionization GOS inevitable. The proximity of the calculation to the experimental GOS of the summary scattering indicates that the result of the RPAE calculation actually includes the contribution of the multiple ionization of the atom.

As to the double ionization, in view of the absence of

any calculations of $K \neq 0$ at the present time, the experimental results can be discussed only qualitatively. A clearly pronounced property of the double-ionization GOS (Fig. 6) is the presence of a maximum similar to the single-ionization GOS maximum, the positions of the two practically coinciding. In contrast to the case of single ionization, one cannot expect such a behavior of $\partial f^{2+}/\partial E$ before hand, starting only from the conservation laws. We have here in the final state three free electrons, and the conservation laws admit of many variants of the distribution of the energy and of the momentum among them. The actual dependence of $\partial f^{2+}/\partial E$ on K is determined by the concrete dynamics of the process of the double ionization. The experimentally observed similarity between the shapes of the $\partial f^{2+}/\partial E$ and $\partial f^+/\partial E$ curves as functions of K corresponds to a double-ionization mechanism such that the interaction of the incident electron with the target proceeds in the same manner as in single ionization, and the energy and momentum transferred in the collision become redistributed among the removed electrons (Fig. 4c and 4d).

Let us examine this question in greater detail, assuming that only two atomic electrons take part in the process. The complete geometric similarity between the $\partial f^{2+}/\partial E$ and $\partial f^+/\partial E$ curves as functions of K would correspond, for example, to double ionization in two stages—knock-out of one electron with subsequent sharing of the energy and momentum by this electron and the second removed electron. Corresponding to this “internal inelastic scattering” is graph d (Fig. 4) under the condition that the intermediate state e'' is real. In addition double ionization via virtual intermediate states are possible (Figs. 4c and 4d at $j = i$). The connection between the energy and the momentum in these relations is not rigid, and can apparently lead to a broadening of the $\partial f^{2+}/\partial E$ curve compared with $\partial f^+/\partial E$. This is precisely what we observed in the experimental results for He.

For Ne and Ar, however, the situation is reversed—the $\partial f^{2+}/\partial E$ curves have a narrower and higher (relative to the value at $K = 0$) maximum than $\partial f^+/\partial E$. The qualitative difference between He, on the one hand, and Ne and Ar, on the other, is apparently connected with the presence in the latter case of two electron subshells. This makes it possible to include the mechanism of double ionization via a virtual Auger transition. In this case one ns electron is initially removed from the atom (Fig. 4c, $j = ns$), after which the produced vacancy is

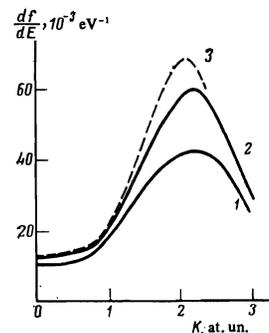


FIG. 7. Generalized oscillator strengths of Ar as functions of the momentum transfer K at $E = 85$ eV. 1—single ionization (experiment), 2—summary inelastic scattering (experiment), 3—single ionization (RPAE calculation).

filled by removal of two np electrons from the atom. This mechanism differs from the usual Auger process, which in this case is energywise forbidden because it proceeds via a virtual intermediate state. According to the data of Chang *et al.*,^[11] this mechanism plays a noticeable role in double photoionization of neon. In the case of argon, the appreciable contribution of this mechanism to double ionization is pointed out by the fact that the shape of the experimental double-ionization GOS curves, as a function of the momentum transfer (Fig. 6c) occupies a position intermediate between the shapes of the calculated single-ionization GOS curve and the curve calculated for a single ionization with removal from the atom of only a $3s$ electron. This intermediate position corresponds to a comparable magnitude of the true mechanism of double ionization—a process connected with the virtual removal of a $3s$ electron from the atom and a “jolt” within the outer shell, which is produced when any of its electrons is removed. The last mechanism, according to Carlson’s estimates^[15] yields 25% doubly-charged ions in the photoionization of the outer shell of Ar.

In connection with the discussion of the role of the virtual Auger process, it is of interest to note that it is possible to associate with this process the result obtained in an experiment^[26] with simultaneous registration of the knock-out and scattered electrons. In this experiment, a maximum of the cross sections was observed when the energy transferred to the Ar atom was 35–50 eV higher than the kinetic energy of the knocked-out electrons, corresponding to the formation of two holes in the $3p$ subshell. At the same time, as shown by analysis in^[26], the correlation of the directions of the emission of the knocked-out electron and the scattered electron corresponds to knocking out of an electron from the s state. Thus, this process can be set in correspondence with graph c of Fig. 4 at $j=3s$ and $i=m=3p$.

Summary. The procedure developed in this paper has made it possible to obtain qualitatively new experimental data—the doubly differential electron inelastic-scattering cross sections resolved into components in accord with the charges of the produced ions. In contrast to the photoabsorption cross sections, these quantities depend not on one but on two parameters, so that additional information can be obtained on the dynamics of the processes and on the role of the interaction between the target electrons. An analysis of the dependence of the GOS on the momentum transfer has led to the following conclusions.

a. The double ionization, like the single ionization, is determined to a considerable degree by the interaction of the incident electron with only one of the target electrons.

b. A comparison of the dependences of the single- and double-ionization GOS on the momentum transfer provides an estimate of the relative role of the different mechanisms of double ionization. The narrower maximum of the double-ionization GOS curve, as compared with the single ionization, is apparently an indication that the virtual Auger process makes a considerable contribution.

c. The increase of the relative yield of doubly charged ions on going to heavier atoms, which is a phenomenon known in the case of photoionization of outer shells of atoms, becomes much stronger when the momentum transfer is increased to values corresponding to the maximum of the GOS.

d. The single-ionization GOS calculated in the RPAE contain effectively also the contribution of the double (and multiple) ionization.

These conclusions, together with the results of our earlier experimental and theoretical investigations of inelastic scattering,^[1,4,5] are in agreement with the following general picture. The inelastic scattering of fast electrons takes place as if the entire energy lost by them were transferred to one atomic electron, regardless of the actual ionization multiplicity. The interactions between the electrons play in this case a double role. First, they influence the distribution of the differential inelastic-scattering cross sections with respect to the energy and momentum transfers—this influence is taken into account in the RPAE method. Second, the interelectron interactions determine the redistribution of the energy transferred in the collision, and consequently the fraction of the cross sections corresponding to ionization of each multiplicity.

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¹Van der Wiel *et al.*^[14] investigated the character of the decrease of this cross section with a scattering angle close to $\phi=0$ at $E_0=10$ keV.

²Recently Van der Wiel (private communication) obtained new oscillator strength values that agree satisfactorily with ours.

³V. V. Afrosimov, Yu. S. Gordeev, V. M. Lavrov, and S. G. Shchemelinin, *Zh. Eksp. Teor. Fiz.* **55**, 1569 (1968) [*Sov. Phys. JETP* **28**, 821 (1969)].

⁴H. F. Wellenstein, R. A. Bonham, and R. C. Ulsh, *Phys. Rev. A* **8**, 304 (1973).

⁵M. Inokuti, *Rev. Mod. Phys.* **43**, 297 (1971).

⁶M. Ya. Amus’ya, N. A. Cherepkov, and S. I. Sheftel’, *Zh. Eksp. Teor. Fiz.* **58**, 618 (1970) [*Sov. Phys. JETP* **31**, 332 (1976)].

⁷M. Ya. Amusia, N. A. Cherepkov, R. K. Janev, and S. I. Sheftel, *Dj. Zivanovic, J. Phys* **B6**, 1028 (1973).

⁸U. Fano and J. W. Cooper, *Rev. Mod. Phys.* **40**, 441 (1968).

⁹R. L. Brown, *Phys. Rev. A* **1**, 341 (1970).

¹⁰F. W. Byron and C. J. Joachain, *Phys. Rev.* **164**, 1 (1967).

¹¹M. Ya. Amusia and M. P. Kazachkov, *Phys. Lett.* **28A**, 27 (1968); M. Ya. Amusia, *In Atomic Physics II*, Plenum, 1971, p. 249.

¹²M. Ya. Amusia, E. G. Drukarev, V. G. Gorshkov, and M. P. Kazachkov, *J. Phys.* **B8**, 1248 (1975); M. S. Yur’ev, *Opt. Spektrosk.* **38**, 9 (1975) [*Opt. Spectrosc. (USSR)* **38**, 4 (1975)].

¹³T. N. Chang, T. Ishihara, and R. T. Poe, *Phys. Rev. Lett.* **27**, 838 (1971); T. N. Chang and R. T. Poe, *Phys. Rev.* **A12**, 1432 (1975).

¹⁴B. L. Schram, *Physica (Utr.)* **32**, 197 (1966); M. J. Van der Wiel *et al.*, *Physica (Utr.)* **42**, 411 (1969).

¹⁵R. H. Hughes and L. D. Weaver, *Phys. Rev.* **132**, 710 (1963).

¹⁶M. J. Van der Wiel and G. Wiebes, *Physica (Utr.)* **53**, 225 (1971); **54**, 411 (1971).

- ¹⁵T. A. Carlson, *Phys. Rev.* **156**, 142 (1967).
- ¹⁶J. A. R. Samson and G. N. Haddad, *Phys. Rev. Lett.* **33**, 875 (1974).
- ¹⁷V. V. Afrosimov, M. Ya. Amusia, Yu. S. Gordeev, M. P. Kazachkov, and S. G. Shchemelinin, *Abstracts of Papers VIII ICPEAC 1*, Belgrade, 1973, p. 415.
- ¹⁸V. V. Afrosimov, I. P. Gladkovskii, Yu. S. Gordeev, I. F. Kalinkevich, and N. V. Fedorenko, *Zh. Tekh. Fiz.* **30**, 1456 (1960) [*Sov. Phys. Tech. Phys.* **5**, 1378 (1961)].
- ¹⁹A. A. Petushkov and A. S. Melioranskiĭ, *Peredovoi nauchno-tekhnicheskii i proizvodstvennyi opyt (Progressive Technical and Manufacturing Practice)*, No. 5, Leningrad, 1960, p. 18.
- ²⁰S. G. Shchemelinin and V. V. Bagaev, *Prib. Tekh. Eksp.* No. 2, 247 (1975).
- ²¹S. G. Shchemelinin and E. P. Andreev, *Zh. Tekh. Fiz.* **45**, 1490 (1975) [*Sov. Phys. Tech. Phys.* **20**, 941 (1975)].
- ²²V. V. Afrosimov, Yu. S. Gordeev, M. H. Panov, and N. V. Fedorenko, *Zh. Tekh. Fiz.* **34**, 1613 (1964) [*Sov. Phys. Tech. Phys.* **9**, 1248 (1965)]; V. V. Afrosimov, Yu. A. Mamaev, M. N. Panov, V. Uroshevich, and N. V. Fedorenko, *Zh. Tekh. Fiz.* **37**, 550 (1967) [*Sov. Phys. Tech. Phys.* **12**, 394 (1967)].
- ²³M. Ya. Amusia and N. A. Cherepkov, *Case Studies in Atomic Physics* **5**, 47 (1975).
- ²⁴L. V. Chernysheva, M. Ya. Amus'ya, and S. I. Sheftel', *Sistema matematicheskogo obespecheniya atomnykh raschetov "ATOM" III. Programma vychisleniya OCO atomov s volnovymi funktsiyami Khartri-Foka i s uchetom mnogoelektronnykh korrelyatsii v odnom perekhode (The "ATOM" III Atomic Computation System Program for the Calculation of Generalized Oscillator Strengths with Hartree-Fock Wave Functions and with Allowance for Many-Electron Correlations in One Transition)*, Preprint FTI-493, Leningrad, 1975.
- ²⁵L. V. Chernysheva, M. Ya. Amus'ya, and S. I. Sheftel', *Sistema matematicheskogo obespecheniya atomnykh raschetov "ATOM" IV. Programma vychisleniya OCO atomov s uchetom mnogoelektronnykh korrelyatsii v dvukh perekhodakh (The "ATOM" IV Atomic Computation Method Program for Calculating the Generalized Oscillator Strengths with Allowance for Many-Electron Correlations in Two Transitions)*, Preprint FTI-495, Leningrad, 1975.
- ²⁶E. Weigold, S. T. Hood, and P. J. O. Teubner, *Phys. Rev. Lett.* **30**, 475 (1973).
- ²⁷J. A. R. Samson, *Adv. in Atomic and Molecular Physics* **2**, 177 (1966).
- ²⁸Y. B. West, G. V. Marr, Y. Hamley, and K. Codling, *Fourth Intern. Conf. on Vacuum-Ultraviolet Radiation Physics*, Hamburg, 1974, p. 176.

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Three-photon ionization of metastable helium atoms

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Direct and resonance three-photon ionization processes are investigated for helium atoms in 2^1S and 2^3S metastable states. The absolute ionization cross sections, the cross section ratios for plane and circularly polarized radiation at given frequency, and the ratios of ionization cross sections from the 2^1S and 2^3S states have been measured for the direct process. All the measured quantities are in good agreement with calculations performed with the aid of time-dependent perturbation theory. A change in the energy of the resonance transition, which is proportional to the radiation intensity, has been found for the ionization process in the presence of the two-photon intermediate 2^1S-6^1S resonance. The origin of perturbations of resonance states is discussed.

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1. INTRODUCTION

The ionization of atoms by high-intensity light has recently attracted the attention of both experimenters and theoreticians. Most of the published investigations have been concerned with the ionization of atoms from the ground state.^[1] The ionization of atoms from excited states is, however, also of considerable interest. In some cases, it enables us to investigate phenomena which cannot be observed in the case of ionization of atoms from the ground state.

Multiphoton transitions in the spectra of noble gases are an example of this. The high ionization energy of these atoms in the ground state ensures that ionization by optical radiation is highly nonlinear ($k=10-20$), so that the field strength must be high and the adiabatic parameter low ($\gamma \approx 1$ ^[1]). Under these conditions, the spectrum of excited states is substantially perturbed by

the field, so that the systematics of levels of the undisturbed atoms is modified and this, among other things, complicates the interpretation of resonances that appear during the multiphoton ionization process.

On the other hand, because of the large energy gap between the ground and excited states in the spectra of noble gases, ionization from the first excited state is much less nonlinear ($k=3-5$). The necessary field strengths are therefore relatively low, and one can investigate in the light field the perturbations of atomic states characterized by the same quantum numbers as in the absence of the field. Since, in this case, the parameter γ is much greater than unity, it is reasonable to expect that perturbation theory should be valid for the theoretical description of ionization from excited states.

In this paper, we shall investigate three-photon ion-