Elastic scattering of slow electrons by atoms

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The total and differential scattering cross sections for 30-eV electrons by He, Ar and Xe atoms are calculated by taking into account multielectron correlations.

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1. In theoretical consideration of the scattering of slow electrons by atoms, the greatest difficulties arise out of the need for account of the exchange between the incident and atomic electrons and the polarization of the atom by the Coulomb field of the incident electron. Many approximate methods have been advanced for solution of the problem of electron-atom collisions.[1] However, they all possess a significant disadvantage—the difficulties of calculation in all these methods increase sharply with increase in the number of electrons.

In the present paper, the apparatus of the quantum theory of many bodies is employed to study the scattering of slow electrons.[2] The self-energy part Σ of the single-particle Green's function is identical with the optical potential of standard scattering theory[3] and the scattering phase shifts are expressed directly in terms of Σ. A method is proposed for calculation of Σ in the simplified random phase approximation with exchange (SRPAE), in which the basic diagrams entering into the random phase approximation with exchange (RPAE) are taken into account. The advantage of such a method of calculation of the scattering phases lies in the absence of any hypothesis of adiabaticity, and also of free parameters. Also of merit is the simplicity of calculation, which allows us to take into account the scattering phases of the slow electrons on practically any atom with filled or half-filled subshells.

The total and differential elastic scattering cross sections of electrons with energies up to 30 eV by He, Ar, and Xe atoms are obtained in the SRPAE. The results of the calculation agree well with the experimental data.

2. We consider the scattering of a slow electron by a neutral atom with Z electrons. In what follows, we neglect the recoil effects and place the atomic nucleus at the coordinate origin; $\mathbf{r}$ is the coordinate of the incident electron, $\mathbf{r}_j$ the coordinates of the atomic electrons. We shall assume that before the collision the atom was in the ground, nondegenerate state. We denote the Hamiltonian of the atom by H_A:

$$H_A\Phi_n=W_n\Phi_n, \tag{1}$$

W_n and Φ_n will be considered known. The energy of the ground state and the wave function are denoted by W_0 and Φ_0, respectively.

The scattering of the electron by the atom is described by the equation[2)]

$$\left[-\frac{\nabla_r^2}{2}-\frac{Z}{r}+\sum_{j=1}^{Z}\frac{1}{|\mathbf{r}_j-\mathbf{r}|}+H_A\right]\Psi=(E+W_0)\Psi \tag{2}$$

(E is the energy of the incident electron). We can expand the wave function Ψ in the complete set of functions Φ_n, and all the information about the elastic scattering is contained in the term of the expansion with $n = 0$:

$$\Psi_{el}=\Psi_0\Phi_0. \tag{3}$$

Our goal is to find an equivalent potential V_{opt} such that Ψ_0 can be obtained from the single-particle equation

$$(-{}^1/_2\nabla_r^2+V_{opt})\Psi_0=E\Psi_0. \tag{4}$$

It is convenient to represent V_{opt} in the following form:

$$V_{opt}=V_{opt}^{(0)}+V_{opt}^{(1)}. \tag{5}$$

The optical potential in the zeroth approximation $V_{opt}^{(0)}$ should be so chosen that the scattering problem in the field $V_{opt}^{(0)}$ has a sufficiently simple solution and the difference $V_{opt}-V_{opt}^{(0)}=V_{opt}^{(1)}$ can at the same time be regarded as a small perturbation. As $V_{opt}^{(0)}$ we take the Hartree-Fock potential of the interaction of the electron with the atom. For this choice of $V_{opt}^{(0)}$ we take the Pauli principle into account, i.e., exchange of the incident electron with the atomic electrons from the very outset. We shall regard the wave functions ψ, which are the solution of the equation

$$(-{}^1/_2\nabla_r^2+V_{opt}^{(0)})\psi=E\psi, \tag{6}$$

as known, and choose them as the basis for finding $V_{opt}^{(1)}$. We represent them in the form

$$\psi=\sum_{lm}(2\pi)^{3/2}i^l e^{i\delta_l}Y_{lm}(\theta_k,\varphi_k)\frac{P_{El}(r)}{(2E)^{1/4}r}Y_{lm}(\theta,\varphi), \tag{7}$$

where θ_k, φ_k and θ, φ are the spherical angles of the directions of the momentum vector of the incident electron and its radius vector. The function $P_{El}(r)$ is the solution of the equation

$$\left(-\frac{1}{2}\frac{d^2}{dr^2}+\frac{l(l+1)}{2r^2}+V_{opt}^{(0)}(r)\right)P_{El}(r)=EP_{El}(r), \tag{8}$$

is normalized to a δ function of the energy, and satisfies the boundary conditions

$$P_{El}(0)=0,\qquad \underset{r\to\infty}{P_{El}(r)}=(2^{1/2}/\pi E^{1/2})^{1/2}\sin[(2E)^{1/2}r-\pi l/2+\delta_l^{(0)}], \tag{9}$$

where $\delta_l^{(0)}(E)$ are the scattering phases in the Hartree-Fock approximation.

In the Born approximation for the potential $V_{opt}^{(1)}$, the correction to the phase $\delta_l^0(E)$ is determined by the expression

$$\Delta\delta_l(E)=-\pi\int P_{El}(r)V_{opt}^{(1)}P_{El}(r)\,dr. \tag{10}$$

The criterion for the applicability of the Born approximation is the condition

$$\Delta\delta_l(E)\ll 1. \tag{11}$$

3. Bell and Squires[3] have shown that the self-energy

part of Σ of the single-particle Green's function G is identical with the optical potential V_{opt}. Actually, the propagation of an additional particle described by the Green's function is determined by its interaction with the atomic electrons. The self energy part is an averaged potential, nonlocal, dependent on the energy and, in general, complex, formed by all the particles of the system.

Similar to (5), we divide Σ into two parts:

$$\Sigma=\Sigma^{\mathrm{HF}}+\Sigma^{(1)} \tag{12}$$

where Σ^{HF} is the self-energy part, found in the Hartree-Fock approximation. Then expression (10) can be written in the following form:

$$\Delta\delta_l(E)=-\pi\int P_{El}(r)\Sigma^{(1)}P_{El}(r)\,d\mathbf{r}. \tag{13}$$

The matrix element of $\Sigma^{(1)}$ is equal to

$$\langle k|\Sigma^{(1)}(\omega)|k\rangle=\sum_{k_1k_2k_3}\int\frac{d\omega_1\,d\omega_2}{(2\pi)^2}\frac{\langle kk_3|V|k_1k_2\rangle}{\omega-\omega_1-E_{k_1}+i\delta(1-2n_{k_1})}\cdot$$
$$\cdot\frac{\langle k_1k_2|\Gamma(\omega,\omega_1,\omega_2)|kk_3\rangle}{[\omega-\omega_2-E_{k_2}+i\delta(1-2n_{k_2})][\omega-\omega_1-\omega_2-E_{k_3}+i\delta(1-2n_{k_3})]}. \tag{14}$$

Here $\langle k_1, k_3|V|k_2k_4\rangle$ are the Coulomb matrix elements:

$$\langle k_1k_3|V|k_2k_4\rangle=\int\varphi_{k_1}^*(\mathbf{r}_1)\varphi_{k_3}^*(\mathbf{r}_2)V(|\mathbf{r}_1-\mathbf{r}_2|)\varphi_{k_2}(\mathbf{r}_1)\varphi_{k_4}(\mathbf{r}_2)\,d\mathbf{r}_1\,d\mathbf{r}_2, \tag{15}$$

$\Gamma(\omega, \omega_1, \omega_2)$ is the amplitude of the effective interaction.[2] The index k denotes the set of four quantum numbers n, l, m and s.

Summation over k includes integration over the continuous spectrum ($k > F$) and summation over the occupied states ($k \leq F$); n_k is the Fermi step: $n_k = 0$, $k > F$; $n_k = 1$, $k \leq F$.

Up to this point, we have made no assumptions as to Γ, and expression (14) for the matrix element of $\Sigma^{(1)}$ is exact. It is known from the theory of the electron gas which processes make a fundamental contribution to Σ in the limiting cases of high and low electron densities.[4] Estimates show[5] that in medium and heavy atoms, one should use the high-density approximation. Calculations of the cross section of photoionization, the polarizability of the atoms, the constants C_6 and C_8 of atomic interaction at large distances, etc.,[5,6] have shown that the application of this approximation in the atom gives excellent results. In the high-density approximation, the largest contribution is made by the infinite series of so-called "lace" diagrams which describe the virtual excitations of one, two, three, etc. particle-hole pairs.[4] In the use of this approximation, which has come to be known as the random-phase approximation (RPA), it is also necessary to take into account the corresponding exchange terms in the atom, inasmuch as the Hartree-Fock approximation and not the Hartree approximation is chosen as the zeroth approximation.

Summation of the graphs by the random-phase method with exchange (RPAE) leads to an equation for the amplitude of the interaction:[5]

$$\langle k_1k_3|\Gamma(\omega)|k_2k_4\rangle=\langle k_1k_3|U|k_2k_4\rangle$$
$$+\Big(\sum_{\substack{k_5\leq F\\k_6>F}}-\sum_{\substack{k_5>F\\k_6\leq F}}\Big)\frac{\langle k_1k_6|\Gamma(\omega)|k_2k_5\rangle\langle k_5k_3|U|k_6k_4\rangle}{\omega-E_{k_5}+E_{k_6}+i\delta(1-2n_{k_5})}, \tag{16}$$

i.e., the matrix of effective interaction in the RPAE approximation depends only on the transferred energy

$$\langle k_1k_3|U|k_2k_4\rangle=\langle k_1k_3|V|k_2k_4\rangle-\langle k_1k_3|V|k_4k_2\rangle. \tag{17}$$

With account of (16), we integrate (14) over ω_2 and, taking the matrix element $\Sigma^{(1)}$ on the mass surface, i.e., for $\omega = E_k$, where E_k is the energy of the incident electron, we obtain for the scattering phase shift in the RPAE approximation

$$\Delta\delta_{l_k}^{\mathrm{RPAE}}(E_k)=-i\pi\sum_l\frac{1}{(2l+1)(2l_k+1)}$$
$$\cdot\sum_{k_1}\Big(\sum_{\substack{k_2>F\\k_3\leq F}}-\sum_{\substack{k_2\leq F\\k_3>F}}\Big)\int\frac{d\omega}{2\pi}\frac{\langle kk_3\|V_l\|k_1k_2\rangle\langle k_1k_2\|\Gamma_l(\omega)\|kk_3\rangle}{[E_k-\omega-E_{k_1}+i\delta(1-2n_{k_1})][\omega-E_{k_2}+E_{k_3}+i\delta(1-2n_{k_2})]}. \tag{18}$$

The reduced matrix elements $\langle\,\|V_l\|\,\rangle$, $\langle\,\|U_l\|\,\rangle$, and $\langle\,\|\Gamma_l\|\,\rangle$ are determined in accord with[6].

In the lowest-order RPAE

$$\langle k_1k_3\|\Gamma_l(\omega)\|k_2k_4\rangle\equiv\langle k_1k_3\|U_l\|k_2k_4\rangle$$

and we can integrate over ω in (18):

$$\Delta\delta_{l_k}^{0}(E_k)=-\pi\sum_l\frac{1}{(2l+1)(2l_k+1)}$$
$$\cdot\Big(\sum_{\substack{k_1k_2>F\\k_3\leq F}}+\sum_{\substack{k_1k_2\leq F\\k_3>F}}\Big)\frac{\langle k_1k_3\|V_l\|k_1k_2\rangle\langle k_1k_2\|U_l\|kk_3\rangle}{E_k-E_{k_1}-E_{k_2}+E_{k_3}+i\delta(1-2n_{k_1})}. \tag{19}$$

Equation (19) is shown graphically in Fig. 1. Here the solid lines with arrows to the right correspond to the propagation of the particle ($k > F$), and those with arrows to the left to the hole ($k \leq F$); the wavy lines correspond to Coulomb interaction.

4. In order that we may use the apparatus of many-body theory to study the scattering of electrons by an atom, the wave functions of the incident and atomic electrons should form a complete orthonormalized set. The wave functions found in the Hartree-Fock approximation satisfy this condition, and the wave functions of the excited states, which we denote by φ^{N+1}, describe the motion of the electron in the field of the neutral atom. In the use of such wave functions in perturbation theory in the interelectronic interaction, difficulties arise that are connected with the logarithmic divergence of the diagonal (in the hole state) Coulomb matrix elements $\langle\epsilon i\|V_l\|\epsilon' i\rangle$ with $l = 0$. In order to avoid these difficulties, we determine the wave function of the particle in the field of a singly charged ion:

$$\Big(-\frac{\nabla^2}{2}-\frac{Z}{r}+\sum_{\substack{j\leq F\\j\neq i}}\int\frac{|\varphi_j(\mathbf{r}')|^2d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}\Big)\varphi_k^N(\mathbf{r})$$
$$-\sum_{\substack{j\leq F\\j\neq i}}\delta(m_{s_j},m_{s_k})\int\frac{\varphi_j^*(\mathbf{r}')\varphi_k^N(\mathbf{r}')d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}\varphi_j(\mathbf{r})=E\varphi_k^N(\mathbf{r})+\sum_{j\leq F}E_{kj}\varphi_j(\mathbf{r}). \tag{20}$$

It has been shown[5] that the use of the functions $\varphi_k^N(r)$ is equivalent to summation of the graphs of the RPAE method that are diagonal in the hole state and that are directed forward in time—see Fig. 2. Here the double line corresponds to propagation of the electron in the field of the ion. We note that the graphs of Fig. 2 are Goldstone graphs and it is impossible to rearrange the order of interaction in time in them (time increases from left to right).

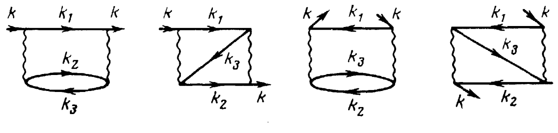

FIG. 1

Allowance for the contribution of multielectron correlations to the scattering phase shift of the electrons in the lowest order of the RPAE approximation by use of the functions φ^{N+1} for the incident electron and the function φ^{N} for the virtual excited states of the atomic electrons will be called the simplified random phase approximation with exchange, SRPAE. In the SRPAE approximation, diagrams with "time reversal," which are taken into account in the RAE, do not appear.

5. We calculated the elastic scattering of electrons with energies up to 30 eV, He, Ar, and Xe atoms. The scattering phase shifts were calculated for the first five partial waves. The contribution of the monopole, dipole and quadrupole components of the Coulomb interaction of the incident electron with the atoms has been taken into consideration. The behavior of the s-, p- and d phases of scattering of the electron by the Xe atom, calculated both in the Hartree-Fock approximation and with account of multielectron correlations in the SRPAE, is shown in Fig. 3. The greatest contribution to the scattering phase shift in the SRPAE is made by the virtual dipole excitations $1s^2 \rightarrow 1s^1\epsilon p$ in He, $3p^6 \rightarrow 3p^5\epsilon d$ in Ar, and $5p^6 \rightarrow 5p^5\epsilon d$ in Xe. In scattering by Ar, the principal contribution to $\Delta\delta$ is made by the virtual excitations of the $3p^6$ subshell, while $3s^2$ is unimportant. Similarly, in elastic scattering of electrons by Xe, the principal contribution to $\Delta\delta$ is made by the virtual excitations of the $5p^6$ subshell, while the contribution of the excitations of the $5s^2$ and $4d^{10}$ subshells is comparatively small. The fact that the shift in the scattering phases is determined essentially by the virtual dipole transition from the upper filled subshell is connected with the circumstance that just this transition makes the largest contribution to the dipole static polarizability,[6] in terms of which the polarization potential is expressed at large distances.[1]

The contribution of the "time backward" diagrams is comparatively small and amounts to 10—15% of $\Delta\delta$.

The scattering phases, calculated both the in the Hartree-Fock approximation and also with the account of multielectron correlations in the SRPAE, satisfy the Levinson theorem, i.e., δ_0, δ_1 and δ_2 tend to π, 0 and 0, respectively, as $E \rightarrow 0$ in the case of scattering from He, to 3π, 2π and 0 for scattering from Ar and to 5π, 4π and 2π for scattering from Xe. This behavior of the phases indicates the absence in these two approximations of discrete levels of bound states of the negative He, Ar or Xe ion. The behavior of the d phases in Ar and Xe is very interesting; with increase in the energy of the incident electron, these phases increase and intersect $\pi/2$ and $5\pi/2$, respectively. This behavior of the d phases indicates the existence of shape resonance in elastic scattering of electrons from Ar and Xe.

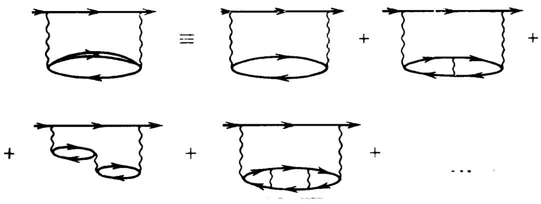

FIG. 2

The phases calculated in the SRPAE are in excellent agreement with those calculated by other authors in the scattering from He, but there is some difference in scattering from Ar, which is especially noticeable at low energies of the incident electron. No calculations of the scattering phases from Xe are known to us.

Knowing the phase δ_0 at small energy and the dipole static polarizability $\alpha_d(0)$ of the atom, we can find the scattering length a from the well-known approximate relation[7]:

$$\delta_0 = -ak - \frac{\pi\alpha_d(0)k^2}{3} - \frac{2}{3}ak^3\alpha_d(0)\ln\left(\frac{\alpha_d(0)k^2}{16}\right). \qquad (21)$$

The values of the scattering length obtained by this formula (the $\alpha_d(0)$ were taken from[6]) for electrons scattered by He, Ar and Xe atoms are given in the Table, where they are compared with the experimental data. It is seen that the results of the calculation of the scattering length in the Hartree-Fock approximation are in poor agreement with the experimental data, and that for Ar and Xe the value of a even has the wrong sign, while, after taking account of multielectron correlations in the SRPAE, the agreement with experiment is very good.

Using the adiabatic approximation, we can estimate how great the difference is between the scattering phases calculated in the SRPAE and those found in the RPAE. Since the principal contribution to the phase shift is made by the dipole transition from the highest subshell occupied, we can assume that

$$\Delta\delta_l^{\mathrm{SRPAE}}/\Delta\delta_l^{\mathrm{RPAE}} \approx \alpha_d^{\mathrm{SRPAE}}(0)/\alpha_d^{\mathrm{RPAE}}(0),$$

where $\alpha_d^{\mathrm{SRPAE}}$ is the static dipole polarizability of the atom, found with use of the function $\varphi^{N(LS)}$.

It was shown earlier[6] that the difference between $\alpha_d^{\mathrm{SRPAE}}(0)$ and $\alpha_d^{\mathrm{RPAE}}(0)$ amounts to no more than 15% for the Ar and Xe atoms. Just such a contribution is made by the diagrams with "time reversal", which are included in the RPAE and not taken into account in the SRPAE. In Figs. 4–6, we have the total elastic scattering cross sections of electrons on He, Ar and Xe atoms both in the Hartree-Fock approximation and with account of multielectron correlations in the SRPAE. The results of our calculations are compared with the experimental data and calculations of other authors. It is seen that the total elastic scattering cross sections of electrons from He, found in the Hartree-Fock approximation and in the SRPAE, are in excellent agreement with the calculations of other authors and with the experimental data.

The total elastic scattering cross section of electrons from Ar, calculated in the Hartree-Fock approximation, differs strongly from the calculation in the SRPAE and from the experimental data. Only after account of multielectron correlations can one describe the Ramsauer minimum. Calculations of the total elastic scat-

Scattering Lengths of Electrons from He, Ar and Xe Atoms

Method of determination	He	Ar	Xe
Level shift [10]	1.12	—1.40	—5.7
Mobility of electrons in the gas [11]	1.19	—1.31	—5.35
Hartree-Fock	1.35	0.85	0.40
SRPAE	1,28	—1,34	—3.65

tering cross section by the method of polarized orbits[9] agree well with out calculation in the SRPAE.

In the Hartree-Fock approximation, we cannot describe the experimentally determined total elastic scattering cross section of electrons from Xe. After account of the multielectron correlations in the SRPAE, the agreement with experiment is very good.

Shape resonances (broad maxima) are seen in the total elastic scattering cross sections of electrons from Ar and Xe in the regions of 1 Ry and 0.5 Ry, respectively, due to resonance in the d wave. After account of the multielectron correlations in the SRPAE, the resonance becomes narrower and shifts in the direction of lower energies. The differential elastic scattering cross section of electrons from the atom is much more sensitive to the approximation used than is the total cross section. Figure 7 gives the results of our calculation of the differential elastic scattering cross section of electrons from He, which are compared with the calculations of others and with experimental data. It is seen that the calculation in the SRPAE is in better agreement with experiment than that in the Hartree-Fock approximation, and is also in agreement with the calculation of other authors.

A calculation of the differential elastic scattering cross section of electrons from Ar is shown in Fig. 8. For an incident electron energy of 0.49 Ry, the differential elastic scattering cross section in the Hartree-Fock approximation has only a single minimum at an angle ~120°, while in the SRPAE there are two minima at ~40° and ~115°, as is confirmed by experiment.

The agreement with the data of Thompson is satisfactory. At a very low energy of the incident electron (E = 0.04Ry), the differential elastic scattering cross section calculated in the Hartree-Fock approximation differs from the experimental cross section by an order of magnitude. The difference between experiment and the differential cross section calculated by the method of polarized orbits is very great. The calculation in the SRPAE is in excellent agreement with the experimental data. The difference between the data of Thompson and the experimental data is connected with the overestimation of the effect of polarization of the atom at low energies of the incident electron.

The results of our calculation of the differential elastic scattering cross section of electrons with energy 0.09 Ry from the Xe atom are shown in Fig. 9. The calculation in the SRPAE is in excellent agreement with the experimental data.

All the above allows us to conclude that the account of multielectron correlations in the SRPAE makes it possible to describe successfully the data of measurements of the total and differential elastic scattering cross sections of low-energy electrons from He, Ar, and Xe atoms.

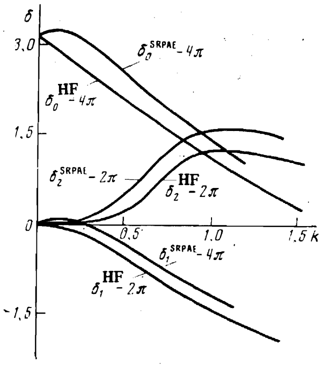

FIG. 3

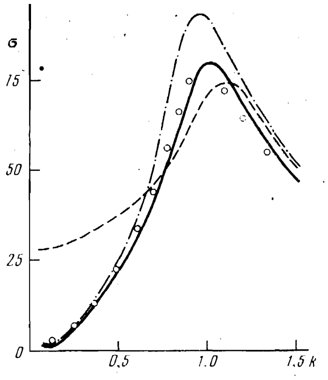

FIG. 5

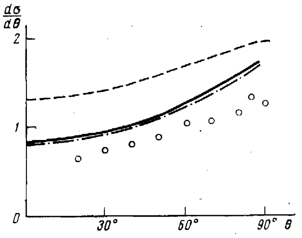

FIG. 7. Differential scattering cross section dσ/dΩ of electrons with energy E = 0.16 Ry from He. Dashed curve–Hartree-Fock, solid curve–SRPAE, dash-dot–method of polarized orbits, [8] ○–experiment [14].

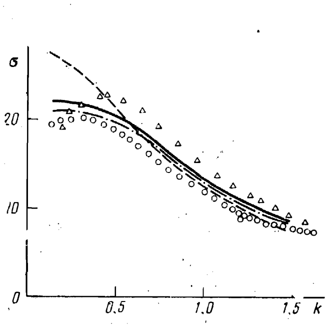

FIG. 4

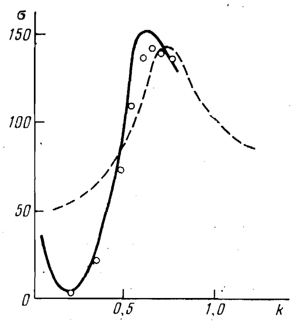

FIG. 6

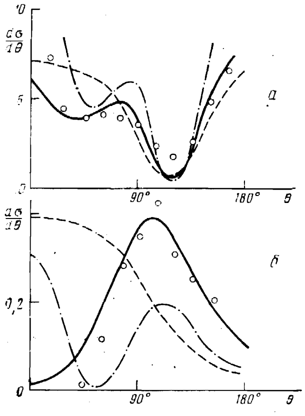

FIG. 8

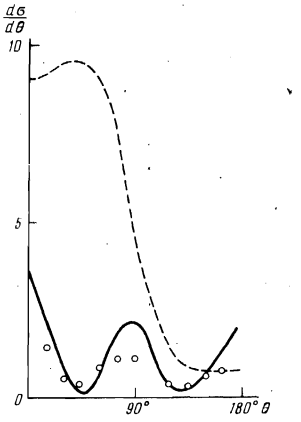

FIG. 9

FIG. 3. s-, p- and d-phases of the scattering of electrons by Xe.

FIG. 4. Total scattering cross section of electrons by He. Dashed curve–Hartree-Fock, solid curve–SRPAE, dash-dot curve–method of polarized orbits, [8] ○–experiment of [12]; △–experiment of [13].

FIG. 5. Total scattering cross section of electrons by Ar. Dashed curve–Hartree-Fock, solid curve–SRPAE, dash-dot–method of polarized orbits, [9] ○–experiment of [12].

FIG. 6. Total scattering cross section of electrons by Xe. Dashed curve–Hartree-Fock, solid curve–SRPAE, ○–experiment of [13].

FIG. 8. Differential scattering cross section dσ/dΩ of electrons a) E = 0.49 Ry, b) E = 0.04 Ry from Ar. Dashed curve (xl/10)–Hartree-Fock, solid curve–SRPAE, dash-dot–method of polarized orbits, [9] ○–experiment [13].

FIG. 9. Differential scattering cross section dσ/dΩ of electrons with energy E = 0.09 Ry from Xe. Dashed curve–Hartree-Fock, solid curve–SRPAE, ○–experiment [13].

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[2] Atomic units are used throughout: $\hbar = m = e = 1$.

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