Study of the current due to photon momentum in atomic gases

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An expression is obtained for the current due to the momentum of a photon in atomic gases. In the one-electron approximation and with inclusion of many-electron correlations we have calculated the currents arising in ionization of gaseous He, Ar, and Xe in the random-phase approximation with exchange. It is shown that in some cases the effect of correlations is quite important.

In discussion of the low-energy photoeffect we usually limit ourselves to the dipole approximation. In this approximation the differential cross section for unpolarized light is described by the following expression[1]:

$$\frac{d\sigma^{nl}(\varepsilon)}{d\Omega_k}=\frac{\sigma^{nl}(\varepsilon)}{4\pi}\left[1-\frac{\beta(\varepsilon)}{2}P_2(\cos\theta_k)\right], \tag{1}$$

where $\sigma^{nl}(\epsilon)$ is the total cross section for the photoeffect of the nl subshell,[2] ϵ is the energy of the photoelectron, θ_k is the angle between the photon and electron momenta, $P_\lambda(\cos\theta_k)$ are Legendre polynomials, and

$$\beta(\varepsilon)=\frac{1}{(2l+1)[lR^2_{l-1}+(l+1)R^2_{l+1}]}\{l(l-1)R^2_{l-1}+(l+1)(l+2)R^2_{l+1} -6l(l+1)R_{l+1}R_{l-1}\cos(\delta_{l+1}-\delta_{l-1})\}, \tag{2}$$

$R_{l\pm1}$ are the dipole matrix elements, and $\delta_\lambda(\epsilon)$ are the phase shifts of the wave functions. The differential cross section (1) does not change its form on replacement of θ_k by $\pi-\theta_k$ and, consequently, is symmetric with respect to the direction of motion of the photon.

Inclusion of higher multipolarities in calculations of this type leads to appearance of asymmetry in the angular distribution of the photoelectrons. Sommerfeld and Schur[3] showed that in hydrogen-like atoms the electrons in the photoeffect are emitted preferentially forward in the direction of motion of the photon. This in turn leads to asymmetry of the emitted-electron momentum distribution function f_k and to the appearance of a current flowing in the direction opposite to the direction of the photon momentum. The effect of appearance of a current upon ionization of impurity centers in semiconductors has been discussed theoretically in terms of a hydrogen-like model in ref. 4.

The present work is devoted to calculation of the current due to the photon momentum in a number of inert gases in the single-particle approximation and with inclusion of many-electron correlations in the random-phase approximation with exchange. (A preliminary report of the results of this work has been published previously.[5]) The magnitude of this current is determined by the quadrupole and dipole matrix elements and also by the phase shifts of the electron wave functions in the continuum. Experimental study of the current would permit an actual direct measurement of the quadrupole transition amplitude at low energies, which is practically impossible by other means. In Ar, Kr, and Xe the study of the current due to photon momentum would permit comparatively accurate determination of the ionization potential of the outer and inner subshells of these atoms and study of the cross section for elastic scattering of electrons by Ar, Kr, and Xe near the Ramsauer minimum.

We will consider the photoelectric effect from the nl subshell of the atom. In the approximation of LS coupling the electron wave functions in the discrete and continuous spectra have the form ($\hbar = m = e = 1$)

$$\psi_{nlm}(\mathbf{r})=\frac{1}{r}P_{nl}(r)Y_{lm}(\theta,\varphi), \tag{3}$$

$$\psi_{\mathbf{k}}^{-}(\mathbf{r})=\frac{(2\pi)^{3/2}}{\sqrt{\varepsilon}}\sum_{\lambda=0}^{\infty}\sum_{\mu=-\lambda}^{\lambda}i^{\lambda}e^{-i\delta_\lambda}Y_{\lambda\mu}^{*}(\theta_k,\varphi_k)Y_{\lambda\mu}(\theta,\varphi)\frac{1}{r}P_{\varepsilon\lambda}(r), \tag{4}$$

where k is the electron momentum, the angles θ_k and φ_k specify the direction of the electron momentum in a coordinate system with a polar axis coinciding with the direction of the photon momentum κ. The radial wave functions of the continuum ($\delta_\lambda(\epsilon)$ are their phase shifts) have been normalized to a δ function of energy and have the following asymptote:

$$P_{\varepsilon l}(r)\to\left(\frac{2}{\pi\sqrt{2\varepsilon}}\right)^{1/2}\sin\left(\sqrt{2\varepsilon}\,r-\frac{\pi l}{2}+\frac{1}{\sqrt{2\varepsilon}}\ln(2r\sqrt{2\varepsilon})+\delta_l\right). \tag{5}$$

The probability of a transition from state (3) to state (4) per unit time induced by an electromagnetic field with vector potential $\mathbf{A}=\mathbf{A}_0\mathbf{e}\exp\{i[\kappa\cdot\mathbf{r}-\omega t]\}=\mathbf{A}(\mathbf{r})e^{-i\omega t}$ is:

$$dW=2\pi\,|\langle\psi_{\mathbf{k}}^{-}(\mathbf{r})\,|\,V(\mathbf{r})\,|\,\psi_{nlm}(\mathbf{r})\rangle|^2\frac{d\mathbf{k}}{(2\pi)^3}\delta(E_{nl}-\varepsilon+\omega), \tag{6}$$

where $E_{nl}<0$ is the ionization potential of the nl subshell and ω is the energy of the absorbed photon. With accuracy to first order in the photon momentum we have

$$V(\mathbf{r})=-\frac{1}{c}\mathbf{A}(\mathbf{r})\mathbf{p}\approx-\frac{A_0}{c}[1+i(\kappa\mathbf{r})](\mathbf{ep}). \tag{7}$$

Here e is the photon polarization vector, p is the momentum operator, and c is the velocity of light.

In Eq. (7) we have not taken into account the electron spin, since in what follows we will limit ourselves to consideration of first-order terms in photon momentum.

Calculation of the matrix element in Eq. (6) for unpolarized light leads to the following expression for the differential cross section for the photoeffect:

$$\frac{d\sigma^{nl}(\varepsilon)}{d\Omega_k}=\frac{\sigma^{nl}(\varepsilon)}{4\pi}\left\{1-\frac{\beta(\varepsilon)}{2}P_2(\cos\theta_k)+\kappa\gamma(\varepsilon)P_1(\cos\theta_k) +\kappa\eta(\varepsilon)P_3(\cos\theta_k)\right\}, \tag{8}$$

where

$$\gamma(\varepsilon)=\frac{3}{5[lR^2_{l-1}+(l+1)R^2_{l+1}]}\left\{\frac{(l+1)}{2l+3}[3(l+2)D_{l+2}R_{l+1}\cos(\delta_{l+2}-\delta_{l+1}) -lD_lR_{l+1}\cos(\delta_l-\delta_{l+1})]-\frac{l}{2l-1}[3(l-1)D_{l-2}R_{l-1}\cos(\delta_{l-2}-\delta_{l-1}) -(l+1)D_lR_{l-1}\cos(\delta_l-\delta_{l-1})]\right\}, \tag{9}$$

$$\eta(\varepsilon)=\frac{3}{5[lR_{l-1}^2+(l+1)R_{l+1}^2]}\left\{\frac{(l+1)(l+2)}{(2l+1)(2l+3)}D_{l+2}[5lR_{l-1}\cos(\delta_{l+2}-\delta_{l-1})\right.$$
$$-(l+3)R_{l+1}\cos(\delta_{l+2}-\delta_{l+1})]-\frac{(l-1)l}{(2l-1)(2l+1)}D_{l-2}[5(l+1)R_{l+1}$$
$$\times\cos(\delta_{l-2}-\delta_{l+1})-(l-2)R_{l-1}\cos(\delta_{l-2}-\delta_{l-1})]+2\frac{l(l+1)}{(2l-1)(2l+3)}$$
$$\left.\times D_l[(l+2)R_{l+1}\cos(\delta_l-\delta_{l+1})-(l-1)R_{l-1}\cos(\delta_l-\delta_{l-1})]\right\}. \quad (10)$$

Here

$$R_{l\pm1}=\int_0^\infty P_{nl}(r)\left[\frac{d}{dr}P_{\varepsilon,l\pm1}(r)\pm\frac{2l\pm1+1}{2r}P_{\varepsilon,l\pm1}(r)\right]dr, \quad (11)$$

$$D_l=\int_0^\infty rP_{nl}(r)\frac{d}{dr}P_{\varepsilon,l}(r)\,dr, \quad D_{l\pm2}=\int_0^\infty rP_{nl}(r)\frac{d}{dr}P_{\varepsilon,l\pm2}(r)\,dr. \quad (12)$$

At $\kappa = 0$, expression (8) goes over into (1). From Eq. (8) it is evident that inclusion of the photon momentum in calculation of the differential cross section for the photoeffect has led to appearance of an asymmetry relative to the direction of motion of the photon, since on replacement of θ_k by $\pi - \theta_k$ the terms with κ in Eq. (8) change sign (it is obvious that this substitution is equivalent to conversion from κ to $-\kappa$). If an electron from a subshell with orbital quantum number $l = 0$ is ejected in the photoeffect, then Eq. (8) takes the form:

$$\frac{d\sigma^{n0}(\varepsilon)}{d\Omega_k}=\frac{3\sigma^{n0}(\varepsilon)}{8\pi}\left[1+2\varkappa\frac{D_2}{R_1}\cos(\delta_2-\delta_1)\right.$$
$$\left.\times\cos\theta_k\right]\sin^2\theta_k=\frac{3\sigma^{n0}(\varepsilon)}{8\pi}[1+b\cos\theta_k]\sin^2\theta_k. \quad (13)$$

In Fig. 1 we have shown the dependence of $d\sigma^{nl}(\epsilon)/d\Omega_k$ on angle θ_k for $l = 0$.

It is interesting to note that in hydrogen-like atoms we have $b > 0$ and this quantity does not change sign with the frequency of the light, which leads to a shift in the maximum of the angular distribution forward in the direction of motion of the photon.[3]

For illustration of the possibility in principle of a shift of the maximum both forward and backwards, let us consider the photoeffect from a rectangular spherical potential well of radius a which for simplicity contains one level with orbital angular momentum equal to zero. In the Born approximation for the ejected electron, the differential cross section for the photoeffect with inclusion of first-order terms in the photon momentum has the form

$$\frac{d\sigma^{n0}(\varepsilon)}{d\Omega_k}\sim\alpha\frac{a_0^2}{(ka)^3\omega}\{\sin^2(ka)-2(\varkappa a)\sin(ka)\cos(ka)\cos\theta_k\}\sin^2\theta_k. \quad (14)$$

It can be seen from (14) that terms with κ change sign with change of the momentum of the photoelectron, which also leads to a shift in the maximum forward or backward (a_0 is the Bohr radius and α is the fine-structure constant).

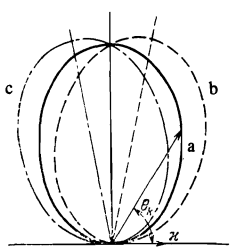

FIG. 1. Dependence of $d\sigma^{n0}(\epsilon)/d\Omega_k$ on angle θ_k; a) in the dipole approximation according to Eq. (1), b) according to Eq. (13) for $b > 0$, c) according to Eq. (9) for $b < 0$.

Let us now investigate the behavior of the free electrons which appear in the gas when it is ionized. Their behavior is described by the Boltzmann kinetic equation, which in the absence of external fields, in the relaxation time approximation, has the form

$$-\frac{f_k-f_k^0}{\tau(\varepsilon)}+\left(\frac{\partial f_k}{\partial t}\right)_{\text{phot}}=0. \quad (15)$$

Here $f_k^{(0)}$ is the equilibrium distribution function of electrons and f_k is the distribution function in the presence of a light wave, $(\partial f_k/\partial t)_{\text{phot}} = N_a W d\sigma^{nl}(\epsilon)d\Omega_k$ determines the rate of change of the asymmetric part of the distribution function due to gas atom ionization processes (W is the photon flux density and N_a is the gas atom concentration), $\tau(\epsilon)$ is the electron momentum relaxation time. Under ordinary real conditions $\tau(\epsilon)$ is determined by elastic scattering of electrons by neutral atoms, and here $\tau(\epsilon) = [N_a\sigma(\epsilon)\sqrt{2\epsilon}]^{-1}$, where $\sigma(\epsilon)$ is the total cross section for elastic scattering of an electron by a gas atom. We will now determine by means of the function f_k the electron velocity projection, averaged over the ensemble, on the photon momentum direction. This quantity is of the form

$$P=\int f_k k\cos\theta_k\,dk. \quad (16)$$

Multiplying Eq. (16) by the electronic charge and taking into account that $\int f_k^0 k\cos\theta_k d\mathbf{k} = 0$, we obtain the following expression for the current density flowing along the direction of propagation of the light:

$$j=-\frac{|e|W}{\sigma(\varepsilon)}\int\frac{d\sigma^{nl}(\varepsilon)}{d\Omega_k}\cos\theta_k\,d\Omega_k. \quad (17)$$

(In Eq. (17) we have used the usual units.)

Substituting Eqs. (1) and (8) into (17), we can verify that in the dipole approximation we have $j = 0$, while in the case (8) the current is different from zero and can change its direction with change of sign of $\gamma(\epsilon)$.

We calculated the current due to photon momentum in He, Ar, and Xe both in the Hartree-Fock one-electron approximation and with inclusion of many-electron correlations in terms of the random-phase approximation with exchange (RPAE). In the latter case, as was shown in ref. 6, the photoionization amplitude is determined by the sum of the infinite series of diagrams shown in Fig. 2. In accordance with this figure, the dipole and quadrupole matrix elements, respectively $\langle k_1|R|k_2\rangle$ and $\langle k_1|D|k_2\rangle$, after taking into account correlations, are determined by the following expressions ($k_1 > F$, $k_2 \le F$, $\omega = E_{k_1} - E_{k_2}$:

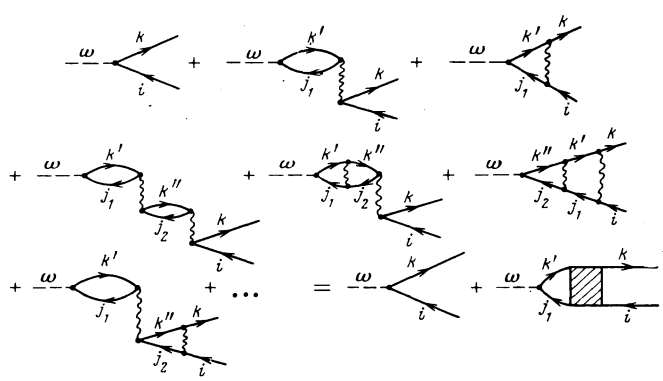

FIG. 2

$$\langle k_1|R|k_2\rangle=\langle k_1|r|k_2\rangle+\Big(\sum_{\substack{k_3\leqslant F\\ k_4>F}}-\sum_{\substack{k_3>F\\ k_4\leqslant F}}\Big)\frac{\langle k_4|r|k_3\rangle\langle k_1k_2|\Gamma(\omega)|k_3k_4\rangle}{\omega-E_{k_4}+E_{k_3}+i\delta(1-2n_{k_4})}, \quad (18)$$

$$\langle k_1|D|k_2\rangle=\langle k_1|q|k_2\rangle+\Big(\sum_{\substack{k_3\leqslant F\\ k_4>F}}-\sum_{\substack{k_3>F\\ k_4\leqslant F}}\Big)\frac{\langle k_4|q|k_3\rangle\langle k_1k_2|\Gamma(\omega)|k_3k_4\rangle}{\omega-E_{k_4}+E_{k_3}+i\delta(1-2n_{k_4})}, \quad (19)$$

where k_i designates the set of four quantum numbers $nlms$,

$$n_{k_4}=\begin{cases}1, & k_4\leqslant F,\\ 0, & k_4>F,\end{cases}$$

F is the Fermi energy, and the interaction amplitude shown in Fig. 2 by the square satisfies the following equation in the RPAE:

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

where $\langle k_1k_3|U|k_2k_4\rangle=\langle k_1k_3|V|k_2k_4\rangle-\langle k_1k_3|V|k_4k_2\rangle$, $\langle k_1k_3|V|k_2k_4\rangle$ are the Coulomb matrix elements.

Calculation of the differential cross section for the photoeffect in terms of the RPAE leads to an expression which formally agrees with Eq. (8), but the following substitutions must be made in Eqs. (9) and (10) in this case:

$$\begin{aligned}&R_{l\pm1}D_{l\pm2}\cos(\delta_{l\pm2}-\delta_{l\pm1})\to(D'R'+D''R'')\cos(\delta_{l\pm2}-\delta_{l\pm1})\\&\quad-(D''R'-D'R'')\sin(\delta_{l\pm2}-\delta_{l\pm1}),\\&|R_{l\pm1}|^2\to R'^2_{l\pm1}+R''^2_{l\pm1},\\&R_{l+1}R_{l-1}\cos(\delta_{l+1}-\delta_{l-1})\to(R'_{l+1}R'_{l-1}+R''_{l+1}R''_{l-1})\cos(\delta_{l+1}-\delta_{l-1})\\&\quad+(R''_{l-1}R'_{l+1}-R'_{l-1}R''_{l+1})\sin(\delta_{l+1}-\delta_{l-1}),\end{aligned} \quad (20)$$

where R′, D′ and R″, D″ are respectively the real and imaginary parts of the matrix elements (18) and (19). We note that in the dipole approximation similar equations were obtained in ref. 7.

Let us dwell now on some details of the current calculation. The continuum wave-function phase shifts entering into Eq. (8) were determined from the equation

$$\sqrt{2\varepsilon}\,r_n-\frac{\pi l}{2}+\frac{1}{\sqrt{2\varepsilon}}\ln(2\sqrt{2\varepsilon}\,r_n)+\delta_l(\varepsilon)=\pi n, \quad (21)$$

where r_n is the distance at which the n-th node of the wave function is found and n is the number of nodes in this distance, the distance r_n being chosen sufficiently large that the phase shift is stabilized ($\gtrsim 100\,a_0$).

The matrix elements (11) and (12) were converted to a form more convenient for the calculations. It is well known that in the case of a local single-particle potential the dipole matrix element (11) can be represented in the form

$$R_{l\pm1}=\frac{1}{\omega}\int_0^\infty rP_{nl}(r)P_{\varepsilon,l\pm1}(r)\,dr. \quad (22)$$

Korolev[8] showed that in cases where it is not necessary to take into account magnetic-dipole transitions, the quadrupole matrix elements also can be converted to the so-called r-form in the following way:

$$\int_0^\infty rP_{nl}(r)\frac{d}{dr}P_{\varepsilon,l\pm2}(r)\,dr=\frac{1}{2\omega}\int_0^\infty r^2P_{nl}(r)P_{\varepsilon,l\pm2}(r)\,dr. \quad (23)$$

In the case of a nonlocal potential the forms r and ∇ give different results, which agree only after taking into account correlations,[6] but in the Hartree-Fock approximation the r form is preferred. It was used by us in the calculations.

The elastic-scattering cross sections $\sigma(\epsilon)$ for Ar and Xe were taken from Ramsauer's article,[9] and for He we used the data of Ramsauer and Kollath.[10] In Ar near the Ramsauer minimum we took the value of $\sigma(\epsilon)$ from Amusia et al.[11] All calculations were carried out for a photon flux density $W = 10^{11}$ cm^{-2} sec^{-1}.

In Fig. 3 we have shown the partial currents arising in ionization of the $3s^2$ and $3p^6$ subshells of Ar, as a function of photon energy. The sharp rise in current near the ionization thresholds of the shells is due to the fact that for photon energies satisfying the relation $\omega + E_{nl} = E_{min}$, where E_{min} corresponds to the Ramsauer minimum, the cross section $\sigma(\epsilon)$ is close to zero and the electrons move in the gas almost without resistance. This fact permits determination of the ionization potentials of the corresponding subshells of the atoms in which the Ramsauer effect is observed. In addition, for a low energy of the photoelectrons, the variation of the matrix elements (22) and (23) and of the phase shifts with energy can be neglected in comparison with the variation of the electron elastic-scattering cross section near the Ramsauer minimum. Therefore the experimental study of the currents in the region of the maxima would permit deduction both of the nature of variation of $\sigma(\epsilon)$ near the minimum and of its magnitude. The effect of correlations in the behavior of the current arising in ionization of the $3s^2$ subshell of Ar turns out to be very important: At the threshold of the $3s^2$ subshell the inclusion of correlations leads to a change in sign of the current with practically no change in its magnitude.

In Figs. 4 and 5 we have shown the total currents arising in ionization of Ar, and Xe, as a function of

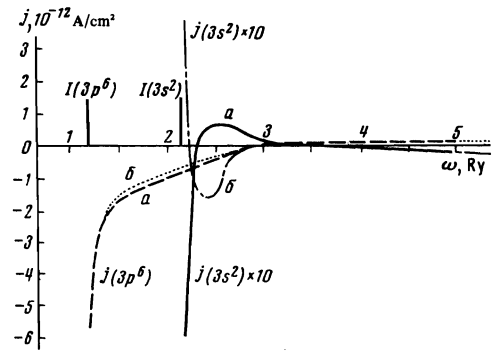

FIG. 3. Dependence of partial currents from $3s^2$ and $3p^6$ subshells of Ar: a) in the one-electron approximation, b) with inclusion of correlations.

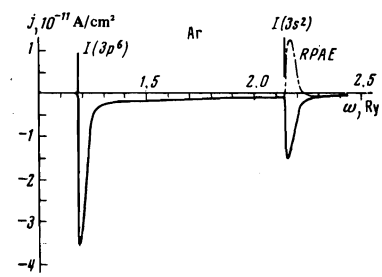

FIG. 4. Total current in Ar.

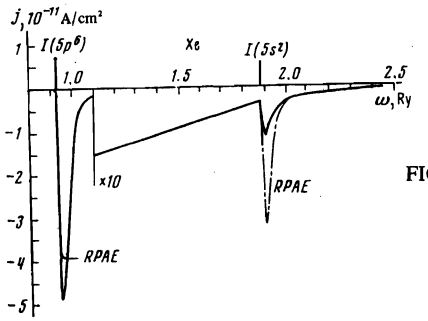

FIG. 5. Total current in Xe.

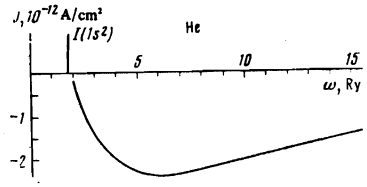

FIG. 6. Current due to photon momentum in He.

photon energy. As the calculations show, the effect of correlations on the behavior of the current arising in ionization of the $5s^2$ subshell of Xe is weaker than in the case of the $3s^2$ subshell of Ar. This is due to the fact that at the ionization threshold of the $5s^2$ subshell of Xe the cosine of the difference in phase shifts, which enters into Eq. (10), turns out to be significantly less than the corresponding quantity in Ar, and therefore the current at the ionization threshold of the $5s^2$ subshell of Xe is less in magnitude than the current at the ionization threshold of the $3s^2$ subshell of Ar. We note that the partial currents in Xe also change their direction with the frequency of the light.

In He, however, as can be seen from Fig. 6, the current does not change sign and flows in a direction opposite to the photon momentum direction. This apparently indicates the hydrogen-like nature of the potential well in He. The effect of correlations on behavior of the current in He turns out to be very weak.

We note that the numbers given in the figures show that the current due to photon momentum is quite accessible for experimental investigation.

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