

Manifestation of the interference of quasimolecular states in radiation polarization when Ca^+ , Sr^+ , and Ba^+ ions collide with inert-gas atoms

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The energy dependences of the total cross sections for the excitation and polarization of the emission of the resonance lines in collisions of Ca^+ , Sr^+ , and Ba^+ ions with inert-gas atoms. It is established that the radiation is strongly polarized in the region of the threshold excitation energies. A new type of interference is observed, causing the experimentally observed oscillations of the polarization functions. A model is proposed for the explanation of the aggregate of the experimental data. Quantitative estimates are obtained of the frequency and amplitude of the oscillations; these estimates agree well with the experimental values.

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The oscillations of the total excitation cross sections in slow collisions of heavy particles have been observed by now for a large number of interacting partners having different structures of their outer electron shells.^[1–3] An oscillatory structure of the energy dependences of the degree of polarization of the excited spectral radiation, uniquely connected with the structure of the total cross section of the corresponding inelastic process, was observed relatively recently.^[3,4] This information has made it possible to reconstruct the quasimolecular particle states that participate in the interference process that is responsible for the oscillations of the total cross sections.^[5]

We report here the results of observation of interference phenomena of a new type, which give rise to a regular oscillating structure of the degree of polarization of the radiation as a function of the ion-atom collision energy. A study of the radiation polarization is one of the steps in our program of systematic investigations of the excitation processes in slow collisions of alkaline-earth ions with inert-gas atoms.^[6,7]

EXPERIMENT

The experiments were performed with a setup whose main units will be described elsewhere. It consists of an ion source with an ion-optical system, a collision chamber, provision for the admission of the gas, an optical system, a spectral instrument, and a system for photoelectric recording of the radiation. The ions produced in a surface-ionization source were focused into a beam that passed through a 127-degree cylindrical electrostatic capacitor and entered the collision chamber. The beam ion current was $(0.1 - 1) \times 10^{-6}$ A with an energy scatter 3–5 eV. The degree of polarization of the optical radiation, observed at a right angle to the direction of the ion beam, was determined with the aid of an Ahrens polarizer. Before reaching the entrance slit of the spectral instrument (MDR-2 or ISP-51), the radiation was depolarized with a Babinet compensator to eliminate the errors connected with the polarizing ability of the monochromator. The optical system made

possible measurements in a wavelength interval above 3800 Å. The radiation leaving the spectral instrument was detected by the method of counting individual photoelectrons of a cooled photomultiplier in three channels of the registration system. The signal accumulated in each of the channels was proportional respectively to the difference between the number of photons polarized parallel and perpendicular to the ion beam direction ($I_{\parallel} - I_{\perp}$), to their sum plus the background count ($I_{\parallel} + I_{\perp} + 2I_b$), and to the background count of the photomultiplier (I_b). The degree of polarization P was defined as

$$P = (I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp}). \quad (1)$$

The gas pressure in the collision chamber was maintained constant in the interval $5 \times 10^{-3} - 5 \times 10^{-4}$ Torr. In this range of pressures and at all values of the ion-beam current, the conditions that the collisions be single were satisfied, and the degree of the radiation polarization remained constant.

Simultaneously with measuring the radiation polarization we investigated the energy dependences of the cross sections for the excitation of the spectral lines. The accuracy of the relative measurements of the excitation functions was 3–4% at the maxima of the curves, and the 90% confidence interval of the measurements of the degree of the polarization is indicated in the figures that illustrate the experimental results.

RESULTS

As established by us earlier,^[6,7] the excitation of the resonant levels of the ions and atoms of the alkaline-earth elements is the dominant process in the collisions of alkaline-earth ions with atoms of inert gases:



(3)

where A^* are the ions Ca^+ , Sr^+ and Ba^+ , and B are the atoms He, Ne, Ar, Kr, and Xe. We have therefore

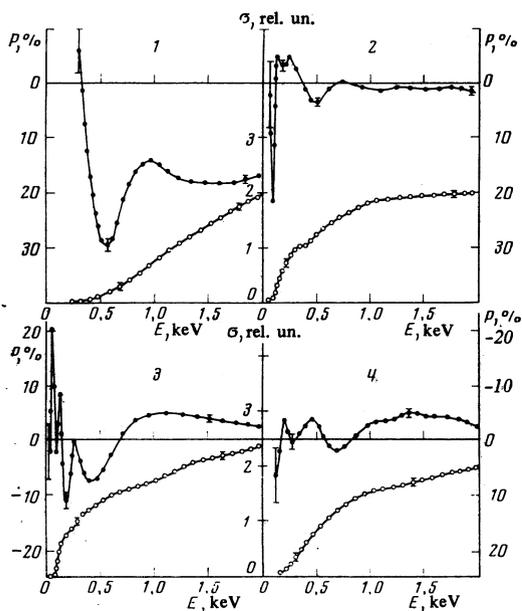


FIG. 1. Energy dependences of the effective excitation cross sections (○) and of the polarization (●) of the resonant radiation of the Ca^+ , Sr^+ , and Ba^+ ions in different processes: 1) $\text{Ca}^+ + \text{Ne} \rightarrow \text{Ca}^{*+} + \text{Ne}$, $\lambda = 3934 \text{ \AA}$ $\text{Ca II}(4^2S_{1/2} - 4^2P_{3/2})$; 2) $\text{Ca}^+ + \text{Ar} \rightarrow \text{Ca}^{*+} + \text{Ar}$, $\lambda = 3934 \text{ \AA}$ $\text{Ca II}(4^2S_{1/2} - 4^2P_{3/2})$; 3) $\text{Ba}^+ + \text{Ne} \rightarrow \text{Ba}^{*+} + \text{Ne}$, $\lambda = 4554 \text{ \AA}$ $\text{Ba II}(6^2S_{1/2} - 6^2P_{3/2})$; 4) $\text{Sr}^+ + \text{Ar} \rightarrow \text{Sr}^{*+} + \text{Ar}$, $\lambda = 4078 \text{ \AA}$ $\text{Sr II}(5^2S_{1/2} - 5^2P_{3/2})$.

paid principal attention to the study of the energy dependences of the polarization of the strong component of the resonant doublets of Ca II ($\lambda = 3934 \text{ \AA}$), Sr II ($\lambda = 4078 \text{ \AA}$), and Ba II ($\lambda = 4554 \text{ \AA}$) ($n^2S_{1/2} - n^2P_{3/2}$) transition, $n = 4, 5$, and 6 respectively for Ca^+ , Sr^+ , and Ba^+ in channel (2). We measured also the polarization functions of the singlet resonance line of the atoms Ca I ($\lambda = 4227 \text{ \AA}$), Sr I ($\lambda = 4607 \text{ \AA}$) and Ba I ($\lambda = 5535 \text{ \AA}$) in channel (3) following bombardment by ions of the atoms Ar , Kr , and Xe . The effective cross section of channel (3) in $\text{A}^+ + \text{He}$ and $\text{A}^+ + \text{Ne}$ collisions is very small, so that in these cases the polarization of the radiation was not investigated, or the obtained data were unreliable. The measurements yielded altogether 24 energy dependences of the degree of polarization of the radiation for 15 different pairs of interacting particles.

Figures 1 and 2 show some typical radiation-polarization functions for channels (2) and (3). To facilitate the analysis, the same figures show the corresponding energy dependences of the effective line-excitation cross sections. The abscissas in each figure represent the energies of the incident ions in the laboratory frame, and the ordinates represent the degree of polarization and the effective cross section in relative units.

Let us examine the main features of the results. As seen from Figs. 1 and 2, the polarization functions are curves with complicated structures. The polarization functions for channel (2) reveal oscillations, while the effective excitation cross section curves are as a rule almost smooth. On going from Ba^+ to Ca^+ , i. e., with decreasing doublet splitting of the final $n^2P_{1/2, 3/2}$ state, the degree of polarization of the emission of the reso-

nance line of the ion decreases, and the structure function $P(E)$ becomes less pronounced. The energy dependences of the effective excitation cross sections and of the degree of polarization of the resonant radiation excited in the charge-exchange process (3) are nonmonotonic functions. However, whereas the structure of the excitation functions manifests itself in most cases in the form of individual singularities, the structure of the polarization functions is distinct in all cases and has an oscillatory character.

In many cases the polarization functions of the resonant emission of the ion exhibit a sharp negative or positive peak. Its energy position corresponds to the region of the rapid increase of the excitation functions at threshold collision energies. It is interesting to note that the same picture was observed in collisions of fast K atoms with Ar , Kr , and Xe atoms,^[8,9] i. e., for partners whose electron shell structures were analogous.

Finally, in processes (2) and (3) the radiation-polarization functions oscillate and reverse sign, while the maximum absolute value of the degree of polarization reaches 35%.

Generalizing the foregoing, we emphasize the following: the singularities in the behavior of the total cross section $\sigma(E)$ give rise to singularities in the behavior of the function $P(E)$, but the oscillatory character of $P(E)$ does not correlate with the structure of the total excitation cross sections. Moreover, as already noted, the oscillations of $P(E)$ are observed also in those cases when $\sigma(E)$ is a smooth function of the collision energy (see Fig. 1, curves 3 and 4).

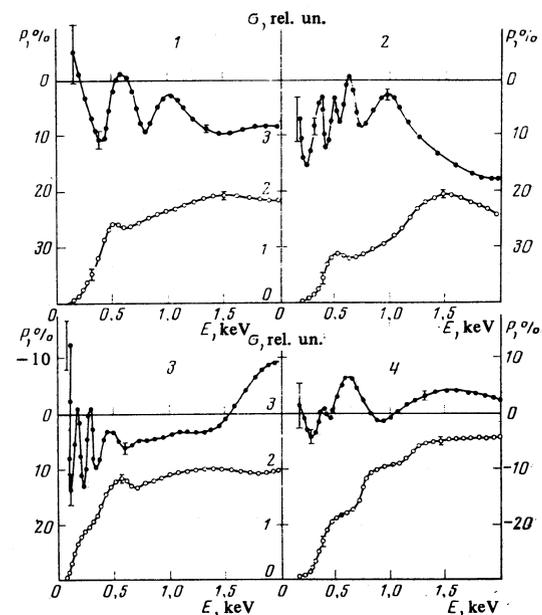


FIG. 2. Energy dependences of the effective cross sections for the excitation (○) and polarization (●) of the resonant singlet line of the atom Ca , Sr , or Ba in the processes: 1) $\text{Sr}^+ + \text{Kr} \rightarrow \text{Sr}^{*+} + \text{Kr}^+$, $\lambda = 4607 \text{ \AA}$ $\text{Sr I}(5^1S_0 - 5^1P_1)$; 2) $\text{Sr}^+ + \text{Ar} \rightarrow \text{Sr}^{*+} + \text{Ar}^+$, $\lambda = 4607 \text{ \AA}$ $\text{Sr I}(5^1S_0 - 5^1P_1)$; 3) $\text{Ca}^+ + \text{Kr} \rightarrow \text{Ca}^{*+} + \text{Kr}^+$, $\lambda = 4227 \text{ \AA}$ $\text{Ca I}(4^1S_0 - 4^1P_1)$; 4) $\text{Ba}^+ + \text{Ar} \rightarrow \text{Ba}^{*+} + \text{Ar}^+$, $\lambda = 5535 \text{ \AA}$ $\text{Ba I}(6^1S_0 - 6^1P_1)$.

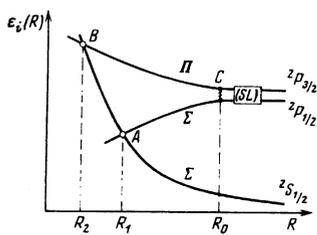


FIG. 3. Schematic behavior of the energies of the quasimolecular terms $\epsilon_i(R)$ as functions of the internuclear distance R .

DISCUSSION OF RESULTS

The excitation of the alkaline-earth metal ion A^+ when scattered by inert-gas atoms B in process (2) is due to population of the excited Σ and Π terms of the molecule $(AB)^*$, which lead to final doublet states of the ${}^2P_{3/2,1/2}$ ion when the particles move apart. The oscillations of $P(E)$ are due in this case to interference of the Σ and Π terms.^[10] Figure 3 shows schematically plots of the energies of the quasimolecular terms $\epsilon_i = \epsilon_i(R)$ against the distance R between the nuclei. The excited Σ and Π terms cross the ground Σ term in the internuclear-distance region $R = R_{1,2}$ in which the terms are predominantly populated. When the colliding particles move apart in the region $R \sim R_0$, mixing of the excited states of the quasimolecule $(AB)^*$ can be produced by the rotation of the internuclear axis. After passing through the remote nonadiabaticity region $R \sim R_0$, a realignment of the electronic state into excited states ${}^2P_{3/2,1/2}$ of the ion takes place.¹⁾ The population of the excited Σ term is due to the radial motion of the colliding particles and takes place in accordance with the Landau-Zener scheme in the vicinities of the pseudo-intersection point $R = R_1$. For the Π state of the molecule, in view of the invariance of the electronic part of the Hamiltonian to reflection in the scattering plane, only the even combination $\varphi^- = [\varphi_{+1} - \varphi_{-1}]/\sqrt{2}$ is populated ($\varphi_{\pm 1}$ are the wave functions of the excited Π term of the quasimolecule with projection $m = \pm 1$ of the orbital angular momentum). When the particles move apart, after passing through the population regions $R = R_{1,2}$, the wave function of the excited state takes the form

$$\Psi = \left[b(\Pi) \frac{\varphi_1 - \varphi_{-1}}{\sqrt{2}} + a(\Sigma) \varphi_0 \right] \begin{pmatrix} d_{\mu} \\ d_{-\mu} \end{pmatrix}, \quad (4)$$

where φ_0 is the wave function of the excited Σ state of the quasimolecule, $b(\Pi)$ and $a(\Sigma)$ are the amplitudes of the population of the excited Π and Σ terms, and d_{μ} are the amplitudes of the spin state of the valence electron with spin projection $\mu = \pm \frac{1}{2}$ on the beam direction.

After the quasimolecular terms are populated, according to the proposed scheme, the system evolves adiabatically up to internuclear distances $R = R_0$. The amplitudes of the wave functions of the states interacting in the vicinities of $R = R_0$ are coupled by a transition matrix whose most general form, based only on the unitarity property, is

$$T = \begin{pmatrix} s & g \\ -g & s \end{pmatrix} = \begin{pmatrix} (1-p)^{1/2} e^{-i\eta_0} & p^{1/2} e^{i\eta_0} \\ -p^{1/2} e^{-i\eta_0} & (1-p)^{1/2} e^{i\eta_0} \end{pmatrix}, \quad (5)$$

where p is the probability of the transition of the quasi-

molecule $(AB)^*$ into another quasimolecular state on passing through the region $R = R_0$, while η_0 and ξ_0 are the phases of the elements of the transition matrix T .

The nonadiabaticity region at $R = R_0$ can be due to the crossing of the Σ and Π terms, when the long-range and exchange interactions, which lift the degeneracy of these states at large R , are of opposite sign. Since the angular velocity of the rotation of the internuclear axis is practically constant in the transition region $R \sim R_0$, the transition probability p takes the form

$$p = \frac{2\pi v}{\Delta F R_0^2} \left(\frac{\rho}{R_0} \right)^2, \quad (6)$$

where ΔF is the difference between the slopes of the terms at $R = R_0$, and v is the collision velocity. In the derivation of (6) we took account of the fact that the population of the vacant terms takes place in a region of internuclear distances substantially smaller than R_0 .

The remote nonadiabaticity region can result also from an abrupt mutual approach of the Σ and Π terms in the vicinities $R = R_0$, when the excited Σ and Π states are practically degenerate for the internuclear distances $R > R_0$. In this case the transition probability is given by

$$p = \rho^2 / R_0^2, \quad (7)$$

where ρ is the impact parameter. The transition probabilities p given by (6) and (7) are small, since $\rho/R_0 \ll 1$.

The expression for the wave function of the system directly after its passage through the nonadiabaticity region is written in the form

$$\Psi = \left[(s'b + ga) \frac{\varphi_1 - \varphi_{-1}}{\sqrt{2}} + (sa - g'b) \varphi_0 \right] \begin{pmatrix} d_{\mu} \\ d_{-\mu} \end{pmatrix}, \quad (8)$$

where

$$b = b(\Pi) \exp \left[-i \int_{t_1}^{t_0} \epsilon_2(R) dt \right], \quad (9)$$

$$a = a(\Sigma) \exp \left[-i \int_{t_1}^{t_0} \epsilon_1(R) dt \right].$$

Further separation of the particles is accompanied by a restructuring of the electronic states into doublet ion states ${}^2P_{3/2,1/2}$. The cross sections $\sigma_{3/2m}$ for the population of the magnetic sublevels of the strong component of the doublet are then given by²⁾

$$\sigma_{\eta_0 \pm \eta_0} = 1/4 \sigma_1 + 1/2 \Delta \sigma \cos \varphi,$$

$$\sigma_{\eta_0 \pm 1/2} = 1/12 [\sigma_1 + 4\sigma_0] - 1/2 \Delta \sigma \cos \varphi, \quad (10)$$

where

$$\Delta \sigma = 2\pi \int_0^{R_0} |a||b| [p(\rho)]^{\eta_0} \rho d\rho \quad (11)$$

is the amplitude of the interference part of the cross section, while σ_1 and σ_0 are the cross sections for the population of the quasimolecular terms:

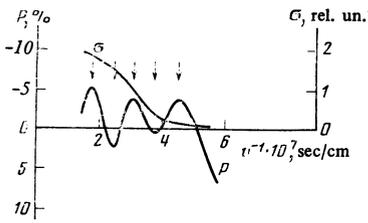


FIG. 4. Excitation cross section and degree of polarization of the ${}^2S_{1/2}-{}^2P_{3/2}$, radiation excited in $\text{Sr}^+ + \text{Ar}$, collisions vs. the reciprocal collision velocity v^{-1} . The arrows indicate the positions of the extrema of the function $P(E)$.

$$\sigma_0 = 2\pi \int_0^{R_1} |a|^2 \rho \, d\rho, \quad \sigma_1 = 2\pi \int_0^{R_1} |b|^2 \rho \, d\rho. \quad (12)$$

The phase φ of the oscillations is

$$\varphi = \frac{1}{v} \frac{\Delta \varepsilon \Delta \bar{R} + \alpha_0}{\Delta \varepsilon \Delta \bar{R} + \alpha_0}, \quad (13)$$

where $\Delta \varepsilon \Delta \bar{R}$ is the oscillation frequency and is equal to the area of the "loop" ABC (see Fig. 3), and α_0 is the initial phase of the oscillations.

The cross sections for the excitation of the magnetic sublevels of the weak component with $J = 1/2$ are

$$\sigma_{\pm 1/2} = 1/2 [\sigma_0 + \sigma_1]. \quad (14)$$

The total cross sections for the excitation of the strong and weak components of the doublet, $\sigma_{3/2}$ and $\sigma_{1/2}$, are obtained by summing of the cross sections of the population of all the magnetic sublevels of the corresponding component:

$$\begin{aligned} \sigma_{3/2} &= \sum_{M} \sigma_{\pm 3/2} = 2/3 (\sigma_1 + \sigma_0), \\ \sigma_{1/2} &= \sum_{M} \sigma_{\pm 1/2} = 1/3 (\sigma_1 + \sigma_0). \end{aligned} \quad (15)$$

As seen from formulas (15), the cross sections for the population of the doublet components are proportional to the statistical weights of the states $J = 3/2$ and $J = 1/2$, as is observed in experiments at sufficiently high collision velocities.^[7]

One of the features of the proposed model is that the total cross section for the population of the strong component of the doublet does not contain the interference terms that are present in the excitation cross sections of the different magnetic sublevels (see formula (10)). It can therefore be concluded that the interference is between quasimolecular terms that populate different magnetic sublevels of the strong component of the doublet. The degree of polarization of this component is expressed in terms of the cross sections for the population of the excited Σ and Π terms:

$$P = \bar{P} + \Delta P = 3 \frac{2\sigma_0 - \sigma_1}{10\sigma_0 + 7\sigma_1} - 18 \frac{\Delta \sigma}{10\sigma_0 + 7\sigma_1} \cos \varphi. \quad (16)$$

As seen from (16), the dependence of the degree of polarization of the radiation is determined by two terms,

the first of which, \bar{P} , is a function of the ratio of the population cross sections of the Π and Σ states (σ_1/σ_0), which varies smoothly with changing collision energy E . The second term, ΔP , is connected with the interference of the quasimolecular states and is a harmonic function of v^{-1} . The regularity of the oscillations is well confirmed by the presented experimental data. Thus, for example, Fig. 4 shows plots of the degree of the radiation polarizations and of the excitation cross section of the strong component of the doublet $5^2P_{3/2}$ of Sr^+ in collisions of Sr^+ with Ar . The extrema of the function P are equidistant accurate to 10%. The cross section $\sigma_{3/2}$ is a smooth function of v^{-1} , thus indicating complete cancellation of the interference terms of the cross sections of different magnetic sublevels. Using the experimental value of \bar{P} , we can obtain the ratio of the cross sections for the population of the quasimolecular states Σ and Π . For example, in the case considered above we have $\sigma_1/\sigma_0 = 2$ in the investigated energy interval.

For an independent estimate of the frequency and of the amplitude of the interference term ΔP we shall use the asymptotic expressions for the potentials of the exchange interaction and for the long-range action^{[11]:}

$$V_{im} = -\frac{\alpha}{2} \left\langle \frac{R+r}{|R+r|^3} - \frac{2R}{R^3} \right\rangle^2 + 2\pi L |\Phi_{im}(R)|^2, \quad (17)$$

where r are the coordinates of the excited electron, $\Phi_{im}(R)$ is the wave function of this electron at $r=R$, α is the polarizability of the inert-gas atom, and L is the length of the scattering of the electron by the same atom. The averaging in (17) is over the wave function of the excited electron. The interaction (17) is responsible for the splitting of the quasimolecular terms Σ and Π , which are degenerate at $R \rightarrow \infty$. The oscillation frequency for the considered case of the collision of Sr^+ with Ar is

$$\Delta \varepsilon \Delta \bar{R} = \int_{R_1}^{R_2} [V_{11}(R) - V_{10}(R)] dR \approx 0.25 \text{ at. un.}, \quad (18)$$

where $R_1 \approx [\sigma_{3/2}/0.43\pi]^{1/2} \sim 1.4$ at. un. is the characteristic dimension of the population region.^[7] The calculated frequency $\Delta \varepsilon \Delta \bar{R}$ agrees well with the frequency value observed in the experiment: 0.23 at. un. The amplitude of the oscillating part, according to estimates based on (17), reaches 4–5%, which is somewhat higher than the experimental 3–3.5%.

Let us examine the process (3) of charge exchange into the excited state 1P_1 of the atom. Within the framework of the proposed model, the charge exchange can be realized via the excited Π and Σ terms of the quasimolecules, which become mixed in some manner at large internuclear distances. Figure 5 shows the functions of the degree of polarization of the emission of the excited state of $\text{Ca}^*(4^1P_1)$ and of the charge-exchange cross section σ as functions of v^{-1} for $\text{Ca}^+ + \text{Kr}$ collisions. The degree of polarization is a regularly oscillating function with frequency $\Delta \varepsilon \Delta \bar{R} \approx 0.35$ at. un. The population cross section is a smooth function of v^{-1} practically everywhere, except the v^{-1} region $(1.5 - 2.4) \times 10^{-7}$ sec/cm, where the regularity of the oscil-

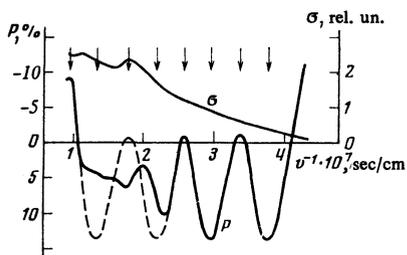


FIG. 5. Excitation cross section σ and degree of polarization P of the emission of the excited atom Ca (4^1P_1) in Ca⁺ + Kr collisions. The upper arrows indicate the positions of the extrema of the functions in the v^{-1} scale. The dashed line shows the approximate oscillations of the polarization function $P(E)$.

lations and the simultaneous appearance of a spike in the charge-exchange cross section corresponds to the opening of new inelastic-scattering channels.

To conclude the evaluation of the results, we note that the appearance of sharp peaks in the degree of polarization in the region where the excitation cross section increases steeply can be explained in the following manner. In the region $R=R_{1,2}$, where the splitting of the Σ and Π terms is large, only one of the quasimolecular states becomes predominantly populated, say Π . In this case the absolute value of the degree of polarization is maximal (it reaches 43% according to (16)). With increasing collision energy, $P(E)$ decreases because of the population of the other quasimolecular states. The total cross section of the process then increases steeply. On the other hand, in the immediate vicinity of the threshold of the process, strong scattering of the colliding particles takes place and leads to depolarization of the radiation.

Summarizing, we note that the observed oscillations of the polarization functions are the consequence of the interference of the quasimolecular terms, that lead to one and the same excited state of the ion (atom). According to the proposed model, the interference terms are present only in the cross sections for the population of the magnetic sublevels of the excited state of the particle, and become cancelled out in the total excitation cross section. Interference phenomena of this type have

been observed here in quasimolecules for the first time, and differ from the heretofore known total-cross-section oscillations due to interference of terms leading to finite states of different particles.^[3,4] The observed oscillations of the degree of polarization were in that case a reflection of oscillations of the total cross sections of the corresponding inelastic processes.^[5] As seen from our results, the presence of a structure of the total excitation cross section is only the sufficient condition for the appearance of a structure in the $P(E)$ dependence. Owing to interference, the degree of polarization can have an autonomous structure.

In conclusion, the authors are deeply grateful to I. P. Zapesochnyi for support and interest, as well as to M. Ya. Amus'ya and S. V. Bobashev for useful discussions.

¹We note that the fine structure of the quasimolecular terms can be neglected in the considered collision-velocity interval.

²Formulas (10)–(14) were derived after averaging over the spin states of the incident particles $|d_\mu|^2 = 1/2$, $d_\mu^* d_{-\mu} = 0$.

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