[2]R. L. Kerber, G. Emanuel, and J. S. Whittier, Appl. Opt. **11**, 1112 (1972).
[3]V. I. Igoshin, Trudy FIAN SSSR 76, 117 (1974).
[4]S. N. Suchard, R. L. Kerber, G. Emanuel, and J. S. Whittier, J. Chem. Phys. 57, 5065 (1972).
[5]Hao-Lin Chen, R. L. Taylor, Jack Wilson, Paul Lewis, and Walter Fyfe, J. Chem. Phys. 61, 306 (1974).
[6]G. K. Vasil'ev, E. F. Makarov, A. G. Ryabenko, and V. L. Tal'roze, Zh. Eksp. Teor. Fiz. 68, 1241 (1975) [Sov. Phys. JETP **41**, 617 (1975)].
[7]J. C. Polanyi and K. B. Woodall, J. Chem. Phys. **57**, 1574 (1972).
[8]G. K. Vasil'ev, V. B. Ivanov, E. F. Makarov, A. G. Ryabenko, and V. L. Tal'roze, Dokl. Akad. Nauk SSSR **215**, 120 (1974).
[9]J. G. Polanyi and J. J. Sloan, J. Chem. Phys. 57, 4988 (1972).
[10]H. K. Shin and A. W. Young, J. Chem. Phys. 60, 193 (1974).
[11]W. G. Tam, Can. J. Phys. **52**, 854 (1974).
[12]J. C. Polanyi, Appl. Opt. Suppl. **2**, 109 (1965).
[13]L. F. Eldreth, A. L. Lovell, and W. F. Herget, J. Opt. Soc. Am. **54**, 715 (1964).
[14]D. F. Smith, J. Mol. Spectrosc. 3, 473 (1959).
[15]H. S. Heaps and G. Herzberg, Z. Phys. **133**, 48 (1952).
[16]W. H. Green and J. K. Hancock, IEEE J. Quantum Electron. **QE-9**, 50 (1973).
[17]R. M. Ozgood Jr., P. B. Sackett, and A. Javan, J. Chem. Phys. 60, 1464 (1974).
[18]K. L. Kompa, J. H. Parker, and G. C. Pimentel, J. Chem. Phys. **49**, 4257 (1968).
[19]G. G. Dolgov-Savel'ev, V. F. Zharov, Yu. S. Meganov, and G. M. Chumak, Zh. Eksp. Teor. Fiz. 61, 64 (1971) [Sov. Phys. JETP **34**, 34 (1972)].
[20]G. G. Dolgov-Savel'ev and A. A. Podminogin, Opt. Spektrosk. **32**, 214 (1972).
[21]N. G. Basov, V. T. Galochkin, V. I. Igoshin, L. V. Kulakov, A. I. Nikitin, and A. N. Oraevsky, Appl. Opt. 10, 1814 (1971).
[22]T. D. Padrick and M. A. Gusinov, Appl. Phys. Lett. **22**, 183 (1973).
[23]S. N. Suchard, R. W. F. Gross, and J. S. Whittier, Appl. Phys. Lett. **19**, 411 (1971).
[24]H. Pummer and K. L. Kompa, Appl. Phys. Lett. **20**, 356 (1972).

Translated by E. Brunner

# Coherent population of quasimolecular states by atomic scattering

S. V. Bobashev and V. A. Kharchenko

*A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences, Leningrad*
(Submitted April 6, 1976)
Zh. Eksp. Teor. Fiz. **71**, 1327–1336 (October 1976)

An analysis is given of the conditions for coherent population of the states of a quasimolecule produced during atomic scattering. It is shown that, within the framework of the model with isolated nonadiabatic interaction regions, the conditions for coherent population of the quasimolecular states improve with decreasing Landau–Stückelberg phases which arise in the region of the population of the quasimolecular terms. Existing experimental data are used to establish conditions for coherent population on the assumption of an effective interaction between these quasimolecular terms throughout the population region due to the rotation of the internuclear axis. Expressions are obtained for the depth of modulation of the total cross sections for different mechanisms of mixing of the interfering terms at large distances. The results are compared with measured total excitation cross sections for collisions between sodium ions and neon atoms.

PACS numbers: 34.50.Hc

A simple model of a collision between two atomic particles, which produces oscillations on the total inelastic cross sections, was discussed previously in[1]. These oscillations are a consequence of interference between two vacant excited states of a quasimolecule which are coherently populated during the collision and interact at large internuclear distances.

According to this model, the ground-state term (term 0) of the system of colliding particles successively crosses the two excited terms 1 and 2 of the quasimolecule for internuclear distances $R=R_1$ and $R=R_2$ (Fig. 1), where the interaction between the ground and excited terms occurs in accordance with the Landau–Zener scheme. As the particles separate, an interaction occurs between the excited terms either due to pseudocrossing or as a result of the close approach at large internuclear distances (for $R=R_3 \gg R_{1,2}$), which leads to the appearance of the interference term $\Delta W$ in the probabilities of population of inelastic scattering channels 1 and 2[1]:

$$\Delta W=2p_1[p_2p_3(1-p_1)(1-p_2)(1-p_3)]^{1/2}\{\cos(\chi+\chi_1-\chi_2)-\cos(\chi+\chi_1)+(1-p_2)\cos(\chi+\chi_2)-p_2\cos(\chi-\chi_2)-(1-2p_2)\cos\chi\}. \quad (1)$$

In this expression, $p_{1,2}$ are the probabilities of conserving the initial electronic state 0 after single crossing of the nonadiabatic regions at $R=R_{1,2}$, $p_3$ is the probability that the system will remain in the same electronic state after nonadiabatic interaction in the region $R=R_3$, and

$$\chi_k=2\varphi_k+\frac{1}{\hbar}\int_{-t_k}^{t_k}(E_k-E_0)\,dt, \quad (2)$$

where

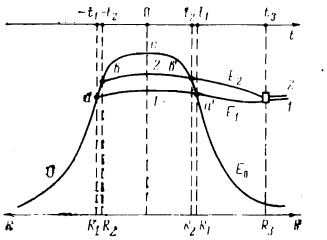

FIG. 1. Schematic representation of the dependence of the energy of quasimolecular terms on the internuclear distance $R$. $E_0 = E_0(R)$ is the energy of the ground-state term, $E_{1,2} = E_{1,2}(R)$ are the energies of the excited terms. The time $t = 0$ corresponds to the classical turning point in the nuclear motion.

$$\varphi_k = \pi/4 + \gamma_k(\ln\gamma_k - 1) - \arg\Gamma(1+i\gamma_k)$$

is the phase due to the Landau–Zener interaction between the terms in the neighborhoods of the points $R = R_{1,2}$,[2] $\gamma_k$ is the Zener parameter for the corresponding nonadiabatic regions,[1)] and

$$\chi = \frac{1}{\hbar}\int_{t_1}^{t_2}(E_2 - E_1)\,dt + \frac{1}{\hbar}\int_{t_2}^{t_1}(E_2 - E_0)\,dt + \varphi_1 - \varphi_2 + \varphi_3. \qquad (3)$$

where the value of the phase $\varphi_3$ is determined by the character of the interaction for $R = R_{1,2}$.

This paper is concerned with the analysis of the population of quasimolecular states under the conditions when the Landau–Stückelberg phases become small in a certain band of collision velocities, which gives rise to an increase in the depth of modulation of the total inelastic cross sections for atomic scattering. We investigate the coherence conditions on the assumption of an effective interaction between the terms of the quasimolecule throughout the entire range of population of vacant excited states of the colliding particles, and analyze the process of population of the quasimolecular terms due to the rotation of the internuclear axis for a close approach between the colliding particles. A comparison is made with experimental results on the oscillations in the total cross sections for collisions between sodium ions and neon.

## 1. POPULATION OF EXCITED STATES IN THE THREE-TERM MODEL

It is clear from the structure of the interference term $\Delta W$ that some of the components are due to the existence of alternative paths which "split" when nonadiabatic regions are traversed as the colliding particles approach one another at the points $a$ and $b$ (Fig. 1). These components are responsible for the Landau–Stückelberg oscillations, the phases of which depend on the impact parameter $\rho$. Such components do not contribute to the oscillating part of the total cross sections. The oscillating part is determined essentially by the component $(1 - 2p_2)\cos\chi$ which appears as a result of interference between alternative paths which "split" only in the nonadiabatic region $R = R_2$ as the particles fly off in opposite directions. There are thus alternative paths ($oabcb'E_2$, $oabcb'a'E_1$) and ($oabb'E_2$, $oabb'a'E_1$). The total cross sections for the corresponding inelastic processes thus retain the interference term $\Delta W$ which does not contain the Landau–Stückelberg oscillations, i.e.,

$$\Delta W = -2p_1[p_2p_3(1-p_1)(1-p_2)(1-p_3)]^{1/2}(1-2p_2)\cos\chi. \qquad (4)$$

Within the framework of the model considered previously,[1] the oscillations in total cross sections appear as a result of the coherent population of states 1 and 2 in regions $R = R_{1,2}$, since the initial phase difference $\eta = \varphi_1 - \varphi_2$ is small and practically independent of the impact parameter $\rho$ ($\varphi_3$ is also independent of the impact parameter). The oscillations in the total cross sections can be observed provided the collision velocity is such that

$$R_1\langle E_2 - E_1\rangle/\hbar v < 1, \qquad (5)$$

where $\langle E_2 - E_1\rangle$ is the average energy difference between terms 1 and 2 for $R \sim R_{1,2}$.[1]

The collisions considered in[1] are those for which the Landau–Stückelberg phases are large, and the associated oscillations are averaged during the integration over all the possible impact parameters. However, for sufficiently high collision velocities $V$, the probable situation is that for which the differences between the adiabatic phases appearing in the population region are small:

$$2R_1\langle E_i - E_k\rangle/\hbar v < 1, \qquad (6)$$

where

$$\langle E_i - E_k\rangle = \int_{\rho/R_1}^{1}\Delta E_{ik}(u)\,[u^2 - \rho^2/R_1^2]^{-1/2}\,u\,du, \qquad (7)$$

and $\Delta E_{ik}(R)$ is the difference between the adiabatic energies of the interacting terms. In this case, the Landau–Stückelberg oscillation phases given by (2) become small and are determined by the quantities $\varphi_{1,2}$. The result of this behavior of the phases is that the terms in the expression for $\Delta W$ given by (1), which contains $\chi_1$ and $\chi_2$, are not averaged during the integration over all the values of the impact parameter and, consequently, provide a contribution to the oscillations in the total cross sections. As the collision velocity increases, $\chi_{1,2} \to \pi/2(\varphi_{1,2} \to \pi/4)$, and the interference term assumes the form

$$\Delta W_c = 4p_1[p_2p_3(1-p_1)(1-p_2)(1-p_3)]^{1/2}p_2\cos\chi. \qquad (8)$$

Comparison of (4) with (8) shows that the amplitude of the oscillations in the population probability and, at the same time, the amplitude of the oscillations in the total cross sections, increase as the Landau–Stückelberg phases vanish, and, as $v \to \infty$, $\Delta W_c/\Delta W = 2$. Thus, the vanishing of the Landau–Stückelberg oscillations as the collision velocity increases leads to an increase in the depth of modulation of the total cross sections. This is

a manifestation of the fact that the conditions for coherent population are satisfied for all the alternative paths leading to the formation of interfering excited quasimolecular states. As the Landau–Stückelberg phases increase, the conditions for coherent population are violated for some of the alternative paths (apart from the $oabcb'E_2$ and $oabcb'a'E_1$, indicated above, and also $oabb'E_2$ and $oabb'a'E_1$), producing a reduction in the depth of modulation of the total cross sections. It is important to note one further feature of term population in the model given in[1]: the depth of modulation cannot be one hundred percent at low velocities $v$ because the alternative path $oaa'E_1$ contributes only to the constant component of the total cross sections and all the interfering terms in the population probabilities due to the existence of this path contain the Landau–Stückelberg phases, i.e., they do not remain in the oscillating part of the total cross sections.

The Landau–Stückelberg phases may be small not only because of an increase in the collision energy but also because of a efficiently small difference between the energies of terms interacting in the population region. In this case, i.e., for small values of $\langle E_i - E_k \rangle$, we may have to face a violation of some of the assumptions lying at the basis of the model[1] and connected with the population of vacant excited terms for $R \leqslant R_1$ in nonadiabatic regions localized in the neighborhoods of the points $a$, $b$, $a'$, $b'$ (Fig. 1). It will be shown below that the conditions for the localization of regions of nonadiabatic interaction in the $R = R_{1,2}$ neighborhoods are not essential for the observation of the oscillatory structure on the total cross sections. When the inequality given by (6) is satisfied, the population of the above quasimolecular states proceeds coherently and independently of the character of the interaction of terms in the population region for $R \leqslant R_1$.

## 2. POPULATION COHERENCE IN THE THREE-TERM MODEL

Let us now consider the population process for the terms of a quasimolecule on the assumption that the energy difference between adiabatic terms in the population region ($R \leqslant R_1$) is sufficiently small, so that the inequality given by (6) is satisfied. In this case, the wave function for the electronic state of the quasimolecule within the framework of the semiclassical approximation has the form

$$\Psi(\mathbf{r}, t) = \sum_{k=0}^{2} b_k(t) \exp\left(-\frac{i}{\hbar}\int^{t} E_k\, dt\right) \psi_k(\mathbf{r}, \mathbf{R}), \tag{9}$$

where $\psi_k(\mathbf{r}, \mathbf{R})$ and $b_k(t)$ are the wave functions and amplitudes for the corresponding states of the quasimolecule, $\mathbf{R} = \mathbf{R}(t)$ is the internuclear position vector, and $\mathbf{r}$ represents the set of electronic coordinates.

In the population region $R = R(t) \leqslant R_1$, the adiabatic amplitudes $b_k(t)$ satisfy the following equations:

$$\frac{db_k}{dt} = -\sum_{n=0}^{2} \left\langle k \left| \frac{d}{dt} \right| n \right\rangle \exp\left[\frac{i}{\hbar}\int_{-t_1}^{t} (E_k - E_n)\, dt\right] b_n, \tag{10}$$

$$\left\langle k \left| \frac{d}{dt} \right| n \right\rangle = \int \psi_k^*(\mathbf{r}, \mathbf{R}) \frac{d}{dt} \psi_n(\mathbf{r}, \mathbf{R})\, d^3r,$$

subject to the initial conditions $b_1 = b_2 = 0$, $b_0 = 1$ when $t = -t_1$.

When the inequality given by (6) is satisfied, the change in the phase factors in (10) during the population time can be neglected (the Landau–Stückelberg phases are small). Under these conditions, the relation between the adiabatic amplitudes is

$$\frac{db_k}{dt} = -\sum_{n=0}^{2} \left\langle k \left| \frac{d}{dt} \right| n \right\rangle b_n. \tag{11}$$

Since all the matrix elements in (11) can be taken to be real, the solution of (11) shows that the phase difference between the adiabatic amplitudes $b_k(t_1)$ immediately after population is identically zero. This ensures that the population of the excited states of the quasimolecule is coherent independently of the nature of the nonadiabatic interaction between the terms. Within the framework of the above analysis, the formation of the quasimolecular amplitudes may be regarded as a "sudden" process, and this is due to the fact that the Landau–Stückelberg phases are small.

As the collision velocity decreases, the inequality given by (6) becomes violated and the corresponding Landau–Stückelberg phases in (10) lead to the appearance of an initial phase difference $\eta = \arg b_1(t_1) - \arg b_2(t_1)$ which, in general, is a function of the impact parameter [$\eta = \eta(v, \rho)$], and the population of the quasimolecular states is no longer coherent.

## 3. TOTAL CROSS SECTIONS FOR INELASTIC PROCESSES FOR A ROTATIONAL INTERACTION OF TERMS

The interaction between adiabatic terms is determined by two factors: the radial motion of the colliding particles and the rotation of the internuclear axis during the collision. The matrix element for the interaction between the adiabatic states can be divided into two terms corresponding, respectively, to the two types of interaction:

$$\left\langle k \left| \frac{d}{dt} \right| n \right\rangle = v_R \left\langle k \left| \frac{d}{dR} \right| n \right\rangle + \frac{1}{\hbar}\omega V_{kn}(R), \tag{12}$$

where $v_R$ is the radial component of the relative collision velocity, $\omega = \omega(\rho)$ is the angular velocity of the internuclear axis, and $V_{kn} = i\langle k | L_x | n \rangle$ is the matrix element of the operator for the component of the electron angular momentum along the direction perpendicular to the plane of rotation of the internuclear axis. For nonadiabatic interactions in the quasimolecule, one of the terms in (12) is zero, in view of the molecular symmetry properties: when $V_{kn} = 0$, the operator $\langle k | d/dt | n \rangle$ connects terms of the same symmetry, whereas, for the interaction between terms of different symmetry ($\Sigma$ and $\Pi$), the radial matrix element becones zero.

Polarization experiments[3,4] and calculations of the degree of polarization[5] for the excitation of a number of NeI lines in the $Na^+ + Ne$ collision have shown that the structure of the excitation functions for the NeI lines is

largely due to interference between the Π electron terms of the quasimolecule. The Π terms can be populated during the rotational interactions with the main Σ term. Analysis of the quasimolecular term diagram for $(NaNe)^*$, reported by Latypov and Shaporenko,[6] enables us to achieve a more detailed description of the population of excited states of the quasimolecule.

If we suppose that the ratio of the matrix elements $V_{20}(R)/V_{10}(R)=\tan\beta$ remains constant in the population region, the solution of (11) for the adiabatic amplitudes $b_k(t_1)$ during the rotational interaction between the terms is

$$b_1(t_1)=\cos\beta\sin\alpha(\rho),\quad b_2(t_1)=\sin\beta\sin\alpha(\rho),\quad b_0(t_1)=\cos\alpha(\rho), \tag{13}$$

where

$$\alpha(\rho)=\frac{1}{\hbar}\int_{-t_1}^{t_1}[V_{10}^2(R)+V_{20}^2(R)]^{1/2}\omega\,dt. \tag{14}$$

The restriction of the population region to internuclear distances $R\leqslant R_1$ is connected with the fact that the ground and excited-state terms diverge widely for $R>R_1$, according to the term diagram given in[6], and the transition probabilities between the terms become small.

In deriving the solution given by (13), we did not take into account the interaction between the Π states of the quasimolecule due to the radial motion of the nuclei of the colliding particles. Since this interaction is small in the region of the effective population of Π terms ($v_R<\omega R$), the coupling of inelastic channels in this region cannot modify substantially the amplitudes of the excited states.

The subsequent development of the amplitudes for the population of excited states of the quasimolecule occurs adiabatically everywhere except for the region of the distant interaction between the terms. After the interaction between the Π terms for $R=R_3$, the population probabilities of the corresponding atomic states 1 and 2 have the form

$$\begin{aligned}W_1&=[p_3\cos^2\beta+(1-p_3)\sin^2\beta]\sin^2\alpha(\rho)+\Delta W,\\ W_2&=[p_3\sin^2\beta+(1-p_3)\cos^2\beta]\sin^2\alpha(\rho)-\Delta W.\end{aligned} \tag{15}$$

The interference part of the population probabilities is

$$\Delta W=[p_3(1-p_3)]^{1/2}\sin 2\beta\sin^2\alpha(\rho)\cos(\Omega/v+\varphi_3). \tag{16}$$

where

$$\Omega=\frac{1}{\hbar}\int_{R_1}^{R_3}(E_2-E_1)[1-\rho^2/R^2]^{-1/2}\,dR$$

is the frequency of the oscillations which is a slowly-varying function of the impact parameter $\rho$ when (5) is satisfied. As already noted, $\varphi_2(v)$ does not depend on the magnitude of the impact parameter, since $\rho/R_3\leqslant R_1/R3\ll 1$.

The total cross sections for inelastic scattering processes connected with the population of atomic states 1 and 2 are obtained by integrating (15) over all the possible values of the impact parameter $\rho\leqslant R_1$:

$$\begin{aligned}\sigma_1(v)&=[p_3\cos^2\beta+(1-p_3)\sin^2\beta]\sigma+\Delta\sigma,\\ \sigma_2(v)&=[p_3\sin^2\beta+(1-p_3)\cos^2\beta]\sigma-\Delta\sigma,\end{aligned} \tag{17}$$

where

$$\Delta\sigma=[p_3(1-p_3)]^{1/2}\sigma\sin 2\beta\cos[\Omega/v+\varphi_3(v)]. \tag{18}$$

In these expressions,

$$\sigma=\sigma_1(v)+\sigma_2(v)=2\pi\int_0^{R_1}\sin^2\alpha(\rho)\,\rho\,d\rho$$

is the total cross section for the population of states 1 and 2 of the quasimolecule. For sufficiently high collision velocities, $\sigma\approx\pi R_1^2/2$ and the total cross section is only slightly dependent on the collision energy,[2)] in agreement with experimental data.[4] We note, moreover, that $\beta$ is a parameter of the quasimolecular terms and is independent of the impact parameter $\rho$.

## 4. DEPTH OF MODULATION OF TOTAL CROSS SECTIONS

The depth of modulation of the total cross sections for inelastic processes when population proceeds only through the excited Π term is determined by two factors, namely, the ratio of cross sections for the population of channels 1 and 2 prior to their interaction in the region $R=R_3$, i.e., $\tan^2\beta=\sigma_2(R_1)/\sigma_1(R_1)$, and the mixing of quasimolecular states in the case of nonadiabatic interactions in the region $R=R_3$, i.e., the quantity $p_3$. The depth of modulation of the cross section for the population of atomic states 1 and 2 is then given by

$$K_1=2[p_3(1-p_3)]^{1/2}\operatorname{tg}\beta/[p_3+(1-p_3)\operatorname{tg}^2\beta], \tag{19}$$

$$K_2=2[p_3(1-p_3)]^{1/2}\operatorname{tg}\beta/[p_3\operatorname{tg}^2\beta+(1-p_3)]. \tag{20}$$

Figure 2 shows the depth of modulation $K_2$ as a function of the reciprocal velocity $v^{-1}$ for different values of the parameter $\beta$ when the region of distant interaction between the terms is described by the Landau–Zener model (Fig. 2a) or the Demkov model (Fig. 2b).

Experimental analysis of the modulation depth of both collision channels can be used together with (19) and (20) to determine the dependence of the quantity $p_3=p_3(v)$ on the collision energy for the region of distant nonadiabatic interaction and the ratio of cross sections for

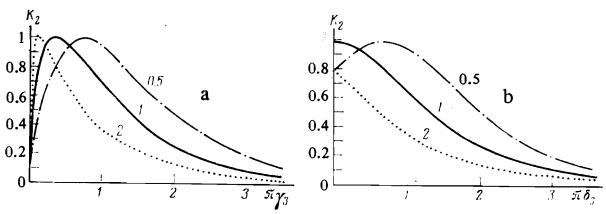


FIG. 2. Modulation depth of the total cross section $\sigma_2(v)$ as a function of the parameters of the distant nonadiabatic region ($\gamma_3$, $\delta_3$) and the ratio of the cross sections for initial population $\tan^2\beta=\sigma_2(R_1)/\sigma_1(R_1)$: a) Landau–Zener model; b) Demkov model. Values indicated against curves are the values of $\tan\beta$.

the population of excited states of the quasimolecule for $R_1 < R < R_3$, i.e., to within their distant interaction.

When the states 1 and 2 are initially populated in the region $R \leq R_1$ with equal probability, i.e., $\sigma_1(R_1) = \sigma_2(R_1)$, the depth of modulation of the cross sections $\sigma_1(v)$ and $\sigma_2(v)$ is the same

$$K_1 = K_2 = K = 2[p_3(1-p_3)]^{1/2}. \quad (21)$$

When the interaction between the terms for $R = R_3$ is described by the Landau–Zener model, the modulation depth is

$$K = 2\exp(-\pi\gamma_3)[1-\exp(-2\pi\gamma_3)]^{1/2}, \quad (22)$$

where $\gamma_3 = H_{12}^2/\hbar v \Delta F$ is the Zener parameter for the region $R = R_3$, $H_{12}$ is the matrix element for the interaction between the pseudocrossing terms 1 and 2, and $\Delta F$ is the difference between the slopes of these terms for $R = R_3$. When the interaction in the region $R = R_3$ is described by the Demkov model, the depth of modulation is

$$K = \mathrm{ch}^{-1}(\pi\delta_3), \quad \delta_3 = (I_2 - I_1)/2(2I_1^{1/2}I_2^{1/2})^{1/2}v, \quad (23)$$

where $\delta_3$ is the Demkov parameter for the region $R = R_3$ and $I_{1,2}$ are the ionization potentials of the corresponding atomic states (in atomic units).

When the initial cross sections for the population of excited quasimolecular states are not equal, the maximum depth of modulation in the first channel is observed for collision velocity $v = v_1$ for which

$$p_3(v_1) = \sin^2\beta = \sigma_2(R_1)/[\sigma_1(R_1) + \sigma_2(R_1)], \quad (24)$$

and, in the second channel, for $v = v_2$ for which

$$p_3(v_2) = \cos^2\beta = \sigma_1(R_1)/[\sigma_1(R_1) + \sigma_2(R_1)]. \quad (25)$$

According to (24) and (25), therefore, the maximum depth of modulation of the cross sections for each of the two inelastic scattering channels is reached for different collision frequencies. Of course, the amplitudes of the interference terms for each of the channels are then equal.

An interesting feature of the behavior of the modulation depth appears when, for large internuclear distances, the quasimolecular states are almost degenerate: $\Delta I_{21} \to 0$. We then have $p_3 = \frac{1}{2}$ for the entire velocity interval, and the modulation depth is the same for both channels

$$K_1 = K_2 = \sin 2\beta = 2[\sigma_1(R_1)\sigma_2(R_1)]^{1/2}/[\sigma_1(R_1) + \sigma_2(R_1)]. \quad (26)$$

When only the Π terms contribute to the total cross sections for the population of interfering atomic states, the observed modulation depth should be quite high (Fig. 2). In fact, measurements of the cross section for the excitation of the λ5852 Å line of NeI by collision with $Na^+$ ions have shown that the modulation depth reaches 80%.[4]

If, in addition to Π terms, the quasimolecular Σ terms contribute to the total cross sections for the population of the corresponding atomic states, the modulation depth for each of the channels is reduced, and the expressions for the modulation depth have the form

$$K_1(v) = \frac{2[p_3(1-p_3)]^{1/2}\,\mathrm{tg}\,\beta}{p_3 + (1-p_3)\,\mathrm{tg}^2\beta + \sigma_1(\Sigma)/\sigma\cos^2\beta}, \quad (27)$$

$$K_2(v) = \frac{2[p_3(1-p_3)]^{1/2}\,\mathrm{tg}\,\beta}{p_3\,\mathrm{tg}^2\beta + 1 - p_3 + \sigma_2(\Sigma)/\sigma\cos^2\beta}, \quad (28)$$

where $\sigma_{1,2}(\Sigma)$ is the cross section for the population of atomic states through the quasimolecular Σ terms.

Figure 3 shows the modulation depth of the total cross section for the excitation of the λ = 736 Å line of Ne during collisions between $Na^+$ and Ne[1,7] as a function of the reciprocal of the collision velocity. The theoretical curve for the modulation depth is based on (27) on the assumption that $\tan\beta = 1$, $\sigma_1(\Sigma) = 3\sigma/2$, which corresponds to the experimentally observed modulation depth of the cross section for the excitation of the λ = 736 Å line of NeI[7] and

$$p_3 = \exp(-2\pi\gamma_3); \quad \gamma_3 = H_{12}^2/v\Delta F = 0.006/v.$$

Thus, the measured dependence of the depth of modulation can be used to determine the parameters of the pseudocrossing for large internuclear distances: $H_{12}^2/F = 0.006$ at. units.

The structure of the $1/v$ dependence of the modulation depth, which has been observed experimentally (Fig. 3), is due, in our view, to weak interference between quasimolecular Σ terms participating in the population of the final atomic states.

There is one other way of obtaining the parameters of the interaction region. It is based on the analysis of the phases of the oscillating parts of the total cross sections [see Eq. (18)]. When the nonadiabatic region at $R = R_3$ appears as a result of the pseudocrossing of terms, the additional phase $\varphi_3(v)$ is determined by the

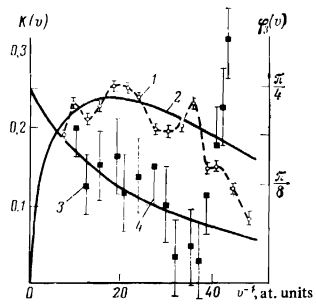


FIG. 3. Modulation depth $K(v)$ and additional phase $\varphi_3(v)$ as functions of the reciprocal collision velocity $1/v$. Curve 1—experimental data for the λ = 736 Å line of NeI excited in the collision $Na^+ + Ne$.[1,7] Curve 2—based on (27) with $\tan\beta = 1$, $\sigma_1(\Sigma) = 3\sigma/2$, $\gamma_3 = 0.006/v$. Curve 3—experimental points obtained by analyzing the interference term in the total cross section for the excitation of the λ = 736 Å line of NeI.[1,7] Curve 4—theoretical value of the Landau–Zener phase $\varphi_3(v)$ for $\gamma_3 = 0.009/v$. The uncertainties indicated correspond to a quadratic deviation with a 90% confidence interval.

parameters of the crossing terms:

$$\varphi_3(v)=(\pi/4)+\gamma_3\ln\gamma_3-\gamma_3-\arg\Gamma(1+i\gamma_3).$$

Figure 3 shows the phase $\varphi_3(v)$ obtained by analyzing the interference term in the total cross sections for the excitation of the $\lambda=736$ Å line of NeI in the case of collisions between $Na^+$ and Ne. For sufficiently high collision velocities, the experimental dependence of the phase on $1/v$ is very close to the behavior of the Landau–Zener phase. This enables us to estimate the parameters of the terms in the distant interaction region, and the result is $H_{12}^2/\Delta F=0.009$ at. units. However, for collision velocities $v\sim 0.03$ at. units, the phase of the interference term begins to increase rapidly. For roughly the same collision velocities, Fig. 3 shows that the modulation depth of the total cross section for the excitation of the NeI lines begins to fall. This suggests that, when $v\sim 0.03$ at. units, the population coherence is violated, i.e., the differences between the adiabatic phases produced in the population region turn out to be of the order of unity, and the condition given by (6) is violated. An analogous reduction in the modulation depth of the total cross sections for inelastic processes is observed with decreasing collision velocity for excitation functions for a number of spectral lines, obtained in experiments on inelastic scattering of various ions by neon atoms.[3,4]

When the interaction at $R=R_3$ occurs as a result of a close approach of the excited quasimolecular terms (Demkov model), the phase $\varphi_3(v)$ becomes a linear function of the reciprocal velocity[2]

$$\varphi_3(v)=\frac{I_2-I_1}{v(I_1I_2)^{1/4}}\frac{\sqrt{2}-\ln(\sqrt{2}+1)}{\sqrt{2}}. \qquad (29)$$

This velocity dependence of the phase $\varphi_3$ leads to an increase in the observed oscillation frequency

$$\Omega'\approx\Omega+0.36(I_2-I_1)/(I_1I_2)^{1/4}\ \text{at. units}.$$

Thus, the measured oscillation frequency is somewhat greater than the area of the "loop" formed by the interfering terms on the diagram showing the dependence of the energy of the quasimolecular terms on the internuclear distance.

Thus, studies of the conditions for coherent population of interfering terms of the quasimolecule performed within the framework of certain model representations provides us with an additional means of interpreting the structural features of both the total cross sections for inelastic processes and the degree of polarization of spectral lines excited during slow ion-atom collisions as a function of energy.

We have confined our comparison with experimental data to inelastic processes in collisions between $Na^+$ ions and Ne because the most complete experimental data are, in fact, available for this ion-atom pair.

The authors are greatly indebted to V. I. Perel' for useful discussions.

[1)] Several terms are missing from the expressions obtained in [1] for the phases $\varphi_k(v)$ because of the approximate evaluation of the phase integrals. Corrected values of $\varphi_k$ are given in [2].

[2)] If we suppose that in the population region $R\leq R_1$ the matrix elements of the rotational interaction are linear functions of $R[V_{k0}(R)=c_kR]$, the total population cross section for states 1 and 2 is

$$\sigma=\frac{\pi R_1^2}{2}-\pi\int_0^{R_1}\cos\left[2b\rho\ln\frac{R_1}{\rho}\right]\rho\,d\rho,$$

where $b=(2c_1v/\hbar)\cos^{-1}\beta$. For high velocities ($b\gg 1$), $\sigma\approx\pi R_1^2/2$, whereas, for small velocities (near the threshold), $\sigma\approx 2\pi b^2\int_0^{R_1}[\rho\ln(R_1/\rho)]^2\rho\,d\rho=\text{const}(E-U_0)$, where $U_0$ is the threshold energy for the inelastic process.

[1] V. A. Ankudinov, S. V. Bobashev, and V. I. Perel', Zh. Eksp. Teor. Fiz. 60, 906 (1971) [Sov. Phys. JETP 33, 490 (1971)].
[2] S. V. Bobashev and V. A. Kharchenko, Preprint No. FTI-462, Leningrad, 1974.
[3] N. H. Tolk, C. W. White, S. H. Neff, and W. Lichten, Phys, Rev. Lett. 31, 671 (1973).
[4] T. Anderson, A. Kirkegaard Nielsen, and K. J. Olsen, Phys. Rev. A 10, 2174 (1974).
[5] E. E. Nikitin, M. Ya. Ovchinnikova, and A. I. Shushin, Pis'ma Zh. Eksp. Teor. Fiz. 21, 633 (1975) [JETP Lett. 21, 299 (1975)].
[6] Z. Z. Latypov and A. L. Shaporenko, Zh. Eksp. Teor. Fiz. 69, 1943 (1975) [Sov. Phys. JETP 42, 986 (1976)].
[7] S. V. Bobashev, Pis'ma Zh. Eksp. Teor. Fiz. 11, 389 (1970) [JETP Lett. 11, 260 (1970)].

Translated by S. Chomet