OSCILLATIONS IN THE TOTAL CROSS SECTIONS FOR INELASTIC PROCESSES DURING ATOMIC COLLISIONS

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A simple model of a collision between two atomic particles, which leads to oscillations in the total cross sections for processes with high resonance defect, is considered. According to Rosenthal's hypothesis, these oscillations are due to interference between two energetically close inelastic channels appearing as a result of term interaction at large distances between the nuclei. It is assumed that two vacant excited levels are successively populated from the ground state in accordance with the Landau-Zener scheme, and then interact as the particles fly apart. The conditions necessary for the appearance of oscillations in the total cross-sections are analyzed, and expressions are obtained for their amplitude, period, and phase. It is shown that, under certain definite conditions, the oscillations are harmonic on the reciprocal velocity scale. The phase of the oscillations is determined by the nature of the excited-term interaction as the particles fly apart. Two variants of this interaction are considered, namely, term crossing and close approach. More complicated models are also considered, including the interaction between a number of terms at large nuclear distances, and it is shown that each closed "loop" formed by these terms corresponds to an oscillation mode in the collision cross sections. The results are compared with some experimental data.

1. INTRODUCTION

IT is well known that the differential cross sections for inelastic processes during collisions between atomic particles are oscillating functions of energy.[1] These oscillations were predicted by Landau and Stückelberg[2] and are due to double transits of the point of pseudocrossing of terms along two alternative paths during the collision process. The oscillatory structure of the total cross sections for the resonant and almost-resonant charge-transfer process has been discussed in the literature in recent years.[3] The observed structure appears as a result of interference between the inelastic and elastic reaction channels, and integration with respect to the impact parameter does not result in the disappearance of the structure only when there is an extremum in the difference between interference terms or "cores" in the scattering potential.[4]

Oscillations in the total cross sections as functions of energy have also been observed for processes involving the excitation of different spectral lines during atomic collisions.[5-8] The essential feature of these excitation processes is the large resonance defect and, therefore, the interaction of the elastic and inelastic channels can occur only near the pseudocrossing of the terms. This leads only to the Landau-Stückelberg-type oscillations in the differential cross sections.

In 1969 Rosenthal[9] suggested that the reason for the oscillations in the total excitation cross sections is the interference between two vacant excited states of the quasimolecule which are coherently excited during collision. According to Rosenthal, the interaction between these states occurs after collision as a result of the crossing of the corresponding quasimolecular terms at large nuclear separations. This hypothesis was used to explain the structure of the excitation functions for the 3^1S- and 3^3S-levels of the $He^+ + He$ process.[6]

One of the present authors[7] has found regular oscillations in the excitation functions for the resonance lines of neon during collisions with Na^+ ions, and suggested that these oscillations may have been the result of interference between the excitation channel and a charge-transfer channel close to it in energy. It is noted in[7] that the term interaction at large nuclear separations may occur as a result of term approach when the colliding particles fly apart. In actual fact, when the terms at infinity are almost degenerate, this approach and the associated modification of the wave functions must necessarily take place, and term crossing is not necessary for term interaction.[1,10]

In the present paper we shall obtain expressions for the inelastic cross sections, taking into account nonadiabatic interactions at large nuclear separations for two inelastic channels. We shall suppose that, when two atomic particles approach one another, the ground-state term for the system crosses successively two vacant excited terms of the quasimolecule. These terms are populated in a coherent fashion, and when the particles fly apart they interact either by pseudocrossing or as a result of approach at large distances. The probability of each of the two inelastic channels turns out to be a harmonically oscillating function of the reciprocal of the relative velocity of the particles. The oscillation frequency is a slow function of the impact parameter. This is so because the impact parameters which are important for the excitation process are much smaller than the distances at which the term

interaction occurs as the particles fly apart. The oscillations therefore persist in the total cross sections as well.

Analysis of more complicated models involving the interaction of a number of terms at large nuclear separations shows that each closed "loop" formed by these terms corresponds to an oscillation mode in the total cross sections.

The term interaction at large nuclear separations should, of course, affect the differential cross sections as well.[1] In contrast to the Landau-Stückelberg oscillations, the oscillations due to this interaction have a much greater period and appear only in the energy dependence of the differential cross sections.

It also follows from the results reported below that if the ground term crosses successively two excited terms then the differential cross section for the excitation of the lower term and for elastic scattering should exhibit the Landau-Stückelberg oscillations corresponding to the upper term, as well as oscillations connected with the Landau-Stückelberg phase difference for two inelastic channels. These oscillations are not connected with term interaction during the separation of the particles. If such interaction does exist, the "difference oscillations" will also appear in the differential cross section for the excitation of the upper term.

2. LANDAU-ZENER TRANSITION IN THE THREE-TERM MODEL

Consider the collision of two atomic particles A and B which results in the appearance of particles A′ and B′ or A″ and B″. We thus have the interaction between the two inelastic processes

$$A+B\to A'+B', \quad A+B\to A''+B''. \tag{1}$$

The excitation of an atom by an ion and charge transfer are examples of this:[7]

$$Na^+ + Ne \to Na^+ + Ne^*, \quad Na^+ + Ne \to Na + Ne^+.$$

Let us suppose that the term scheme for the quasimolecule produced during the collision is of the form shown in Fig. 1. In this figure 0 denotes the ground term of A + B, whereas 1 and 2 indicate the excited terms of A′ + B′ and A″ + B″. As the particles approach one another during the collision process, the energy of the quasimolecule varies along the curve 0. At the pseudocrossing points R_1 and R_2 the excited states 1 and 2 become populated. In region R_3, shown by the rectangle in Fig. 1, these states interact as the particles fly apart. We shall assume that the term interaction occurs near R_1, R_2, and R_3. For other nuclear separations the wave function for the system develops adiabatically and is of the form

$$\psi = b_0(t)\psi_0(R)+b_1(t)\psi_1(R)+b_2(t)\psi_2(R), \tag{2}$$

where $\psi_0(R)$, $\psi_1(r)$, and $\psi_2(R)$ are the wave functions for states 0, 1, 2 of the quasimolecule for a nuclear separation R, and $b_0(t)$, $b_1(t)$, and $b_2(t)$ depend on time only through the phase factor $\exp\{-i\hbar^{-1}\int^t E_i(R)dt\}$, where $E_i(R)$ are the energies of the corresponding states.

We shall solve the problem in two stages. Firstly, we shall find the amplitudes b_1 and b_2 directly after the states 1 and 2 become populated at the point R_1, and then we shall consider their interference due to term interaction for large nuclear separations R_3 as the particles fly apart.

We shall suppose henceforth that the colliding particles move uniformly in a straight line, i.e., the kinetic energy associated with the relative motion of the particles is greater than the potential energy of the interaction between them.

We shall divide the time interval $-\infty < t < \infty$ into a number of subintervals, and investigate the coefficients $b_i(t)$ in Eq. (2) as these regions are traversed. It will be convenient to take the term scheme in the form shown in Fig. 2, where the left-hand side corresponds to the approach and the right-hand side to the separation of the particles. As the particles A and B approach each other ($t < 0$) the ground term 0 of the quasimolecule crosses the two terms 1 and 2 at times t_1' and t_2', where the two terms correspond to two vacant excited states; $t = 0$ corresponds to the turning point. When the particles separate, the pseudocrossings R_2 and R_1 are traversed again. As a result, the system is a superposition of states 0, 1, 2. State 0 need not be considered henceforth. Each pseudocrossing will be treated within the framework of the "two-level" model in accordance with the Landau-Zener scheme, assuming that the pseudocrossing points are sufficiently isolated from each other and are far enough from the turning point.

Following Zener,[11] we can establish the relation between the amplitudes for the states 1 and 2 to the left of the point of intersection $t = t_k$ (which we shall denote by b_1^- and b_2^-) and to the right of this point (b_1^+ and b_2^+):

$$b_1^+ = g_k b_1^- + s_k^* b_2^-, \quad b_2^+ = -s_k b_1^- + g_k b_2^-. \tag{3}$$

In these formulas

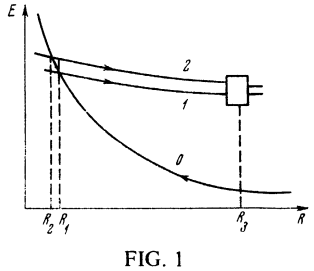

FIG. 1

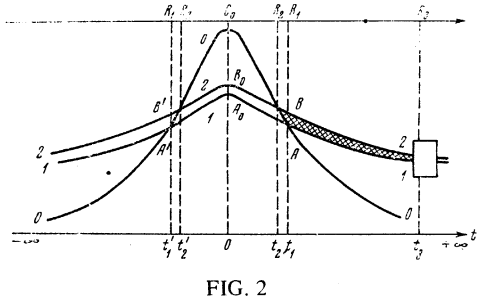

FIG. 2

$$g_k = \exp(-\pi\gamma_k), \tag{4}$$

$$s_k = \frac{\sqrt{2\pi\gamma_k}}{\Gamma(1+i\gamma_k)} \exp\left\{-\frac{\pi}{2}\gamma_k + i\gamma_k \ln\gamma_k + i\frac{\pi}{4}\right\}, \tag{5}$$

where γ_k is the Landau-Zener parameter

$$\gamma_k = V_k^2/\hbar v_R|\Delta F_k|, \tag{6}$$

V_k is the matrix element of the interaction connecting the two intersecting terms, ΔF_k is the force difference corresponding to these terms at the point of intersection R_k, and v_R is the radial relative velocity at this point. To obtain the expressions for the amplitudes of states 1 and 2 directly after the population at R_1, we must successively apply the transformation given by Eq. (3) to all the crossings which occur between $t = -\infty$ and $t = t_1$, inclusive, subject to the initial conditions $|b_0(-\infty)| = 1$, $b_1(-\infty) = b_2(-\infty) = 0$. Moreover, we must, of course, take into account the adiabatic phase change between the crossings.

The system may reach each of the states 1 and 2 in various ways. State 1 can be reached along the three paths $0A'A_0A1$, $0A'B'B_0BA1$, or $0A'B'C_0BA1$, whereas state 2 can be reached along $0A'B'B_0B2$ or $0A'B'C_0B2$. The expressions for the amplitudes are given below, where each of the terms corresponds to a definite path in the order indicated above:

$$b_1(t_1) = s_1^* g_1 \exp\left(-\frac{i}{\hbar}\int_{t_1'}^{t_1} E_1 dt\right) - g_1 s_2^{*2} s_1 \exp\left(-\frac{i}{\hbar}\int_{t_1'}^{t_2'} E_0 dt - \frac{i}{\hbar}\int_{t_2'}^{t_2} E_2 dt - \frac{i}{\hbar}\int_{t_2}^{t_1} E_0 dt\right) - g_1 g_2^2 s_1 \exp\left(-\frac{i}{\hbar}\int_{t_1'}^{t_1} E_0 dt\right), \tag{7}$$

$$b_2(t_1) = g_1 s_2^* g_2 \exp\left(-\frac{i}{\hbar}\int_{t_1'}^{t_2'} E_0 dt - \frac{i}{\hbar}\int_{t_2'}^{t_1} E_2 dt\right) - g_1 g_2 s_2 \exp\left(-\frac{i}{\hbar}\int_{t_1'}^{t_2} E_0 dt - \frac{i}{\hbar}\int_{t_2}^{t_1} E_2 dt\right). \tag{8}$$

We can now write down the expressions for the probability of excitation of states 1 and 2 after a double transit through the pseudocrossing region in the three-term model:

$$\begin{aligned} |b_1(t_1)|^2 &= 2p_1(1-p_1)\{1 - p_2 + p_2^2 - (1-p_2)\cos(\chi_1 - \chi_2) \\ &\quad + p_2(1-p_2)\cos\chi_2 - p_2\cos\chi_1\}, \\ |b_2(t_1)|^2 &= 2p_1p_2(1-p_2)(1-\cos\chi_2). \end{aligned} \tag{10}$$

In these expressions $p_k = g_k^2 = \exp(-2\pi\gamma_k)$ is the probability of remaining in a given state during the k-th pseudocrossing,

$$\chi_k = 2\varphi_k + \frac{1}{\hbar}\int_{t_k'}^{t_k} (E_k - E_0)\, dt, \tag{11}$$

and φ_k is the phase of the coefficient s_k defined by Eq. (5):

$$\varphi_k = \gamma_k \ln\gamma_k + {}^1/_4\pi - \arg\Gamma(1+i\gamma_k). \tag{12}$$

In the special case where one of the terms is absent ($p_2 = 1$ or $p_1 = 1$), Eqs. (9) and (10) reduce to the well-known Landau-Stückelberg expression[2] when the potential energy of the interaction is much less than the kinetic energy of relative motion.

It is clear from Eq. (9) that when the interactions of all three terms are taken into account, the result is that the population probability for the lower term acquires some additional components. In addition to the usual oscillations connected with the phase χ_1 we also obtain oscillations connected with the phase χ_2. The most interesting effect is the appearance of interference oscillations described by the phase difference $\chi_1 - \chi_2$. These additional oscillations appear as a result of interference along $0A'A_0A1$ and $0A'B'B_0BA1$. If the terms 1 and 2 are energetically close to the ground term in the pseudocrossing region, then the differential cross section for inelastic scattering may exhibit low-frequency modulation of the Landau-Stückelberg oscillations. The same modulation will obviously appear in the case of elastic scattering.

Equations (9) and (10) describe the populations of states 1 and 2 which would appear after collision if the terms 1 and 2 did not interact during the separation of the particles. However, in this case, integration with respect to the impact parameter will ensure that there will be no oscillations in the total cross sections. We note that, when Eq. (9) is averaged over the oscillations, the result is identical with Eq. (6) of[12].

3. ALLOWANCE FOR THE INTERACTION DURING PARTICLE SEPARATION

Let us now return to the region $t = t_3$ ($R = R_3$) where, according to the hypothesis formulated above, there is an additional nonadiabatic interaction between the separating particles.

Near the point R_3, and to the left of it, the amplitudes $b_1^-(t_3)$ and $b_2^-(t_3)$ differ from the amplitudes $b_1(t_1)$ and $b_2(t_1)$ given above merely by the phase factors

$$\begin{aligned} b_1^-(t_3) &= b_1(t_1)\exp\left\{-\frac{i}{\hbar}\int_{t_1}^{t_3} E_1(R)\, dR\right\}, \\ b_2^-(t_3) &= b_2(t_1)\exp\left\{-\frac{i}{\hbar}\int_{t_1}^{t_3} E_2(R)\, dR\right\}. \end{aligned} \tag{13}$$

Near the point R_3, and to the right of it, the amplitudes for these states will have different values, namely, $b_1^+(t_3)$ and $b_2^+(t_3)$, each of which is a linear combination of $b_1^-(t_3)$ and $b_2^-(t_3)$.

Let us consider two variants of the interaction, leading to the shift of the states during the separation of the particles.

If we follow Rosenthal[9] and assume that terms 1 and 2 cross at large nuclear separations $R = R_3$ (Fig. 3a), the relation between $b_1^+(t_3)$, $b_2^+(t_3)$ and $b_1^-(t_3)$, $b_2^-(t_3)$ is given by Eqs. (3)–(6).

In the second variant, where the sharp approach of the terms in region R_3 is responsible for the nonadiabatic interaction, we can employ the results of Demkov[10], who used this approach to construct a theory of charge transfer for small resonance defects. The only difference is that, in our case, the system traverses only once the region of term approach (as the particles fly apart).

Following Demkov[10], we shall suppose that the diagonal matrix elements of the Hamiltonian, H_{11} and H_{22}, are constant in the region R_3, whereas nondiag-

onal elements are proportional to $e^{-\lambda R}$. Since the approach occurs at large nuclear separations, we may suppose that R = vt. The point R_3 (and hence t_3) is conveniently chosen so that the nondiagonal element of the Hamiltonian at this point is equal to the half-difference between the diagonal elements. Let us therefore write

$$H_{12} = H_{21} = {}^1/_2 k(H_{22} - H_{11})\exp\{-\lambda(R - R_3)\}, \qquad (14)$$

where k = ±1. The adiabatic terms of the quasimolecule are then of the form

$$E_{2,1} = {}^1/_2(H_{11} + H_{22}) \pm {}^1/_2(H_{22} - H_{11})\,(1 + e^{-2\lambda(R-R_3)})^{1/2}. \qquad (15)$$

The terms approach for R ~ R_3 (Fig. 3b). Their adiabatic wave functions ψ_1 and ψ_2 are modified during the approach in such a way that they become equal to the half-sum and half-difference of the functions prior to term approach. The modification corresponds to a transition from the molecular wave functions to functions for a system of isolated atoms.

Let us now summarize the results of our solution of the Schroedinger equation for the term-approach region. For the appropriate choice of the phases the relation between the wave functions before and after the approach is

$$\psi_1^+ = \frac{1}{\sqrt{2}}(\psi_1^- + \psi_2^-)\exp\left(-i\delta\ln\frac{\delta}{2} - i\theta + i\delta\right),$$

$$\psi_2^+ = \frac{1}{\sqrt{2}}(\psi_2^- - \psi_1^-)\exp\left(i\delta\ln\frac{\delta}{2} + i\theta - i\delta\right), \qquad (16)$$

where θ is the argument of the gamma function $\Gamma({}^1/_2 - i\delta)$ and

$$\delta = (H_{22} - H_{11})/2\hbar\lambda v. \qquad (17)$$

The coefficients $b_1^+(t_3)$, $b_2^+(t_3)$ and $b_1^-(t_3)$, $b_2^-(t_3)$ are then related by Eq. (3), except that g_3 and s_3 are now given by

$$g_3 = (1 + e^{-\pi\delta})^{-1/2}, \qquad s_3 = (1 + e^{\pi\delta})^{-1/2}e^{2i\delta}. \qquad (18)$$

It is clear from these two expressions that when $\lambda \to 0$ (very smooth term approach) we have $s_3 \to 0$ and there are no transitions. For large λ (sharp term approach) the states are fully mixed and $g_3 = |s_3| = 1/\sqrt{2}$.

According to Eqs. (3) and (13), the probabilities of population of the states 1 and 2 after the particle collisions can be written in the form

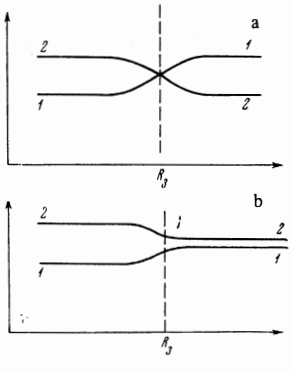

FIG. 3

$$|b_1^+(t_3)|^2 = W_1 + \Delta W, \qquad |b_2^+(t_3)|^2 = W_2 - \Delta W, \qquad (19)$$

where

$$W_1 = |b_1(t_1)|^2 g_3^2 + |b_2(t_1)|^2 |s_3|^2,$$
$$W_2 = |b_1(t_1)|^2 |s_3|^2 + |b_2(t_1)|^2 g_3^2. \qquad (20)$$

The squares of the moduli of $b_1(t_1)$ and $b_2(t_1)$ are given by Eqs. (9) and (10). The interference term ΔW depends on the behavior of terms 1 and 2 in the region between the pseudocrossing R_1 and the interaction point R_3, and is given by

$$\Delta W = 2\,\mathrm{Re}\, s_3 g_3 b_1(t_1) b_2^*(t_1)\exp\left\{\frac{i}{\hbar}\int_{t_1}^{t_3}[E_2(R) - E_1(R)]\,dt\right\}. \qquad (21)$$

We recall that g_3 and s_3 are given by Eqs. (4) and (5) if the terms cross in region R_3, and by Eq. (18) if they approach one another.

Using Eqs. (7) and (8), we obtain

$$\Delta W = 2p_1[p_2p_3(1-p_1)(1-p_2)(1-p_3)]^{1/2}\{\cos(\chi + \chi_1 - \chi_2) - \cos(\chi + \chi_1) + (1-p_2)\cos(\chi + \chi_2) - p_2\cos(\chi - \chi_2) - (1-2p_2)\cos\chi\}, \qquad (22)$$

where $p_3 = g_3^2$. The phases χ_1 and χ_2 are given by Eq. (11) and

$$\chi = \frac{1}{\hbar}\int_{t_1}^{t_3}(E_2 - E_1)\,dt + \frac{1}{\hbar}\int_{t_2}^{t_1}(E_2 - E_0)\,dt + \varphi_1 - \varphi_2 + \varphi_3. \qquad (23)$$

In this expression φ_3 is the phase of s_3 which, in the case of term approach, is equal to $+2\delta$, whereas in the case of crossing it is given by Eq. (12).

4. OSCILLATIONS IN TOTAL CROSS SECTIONS AND DISCUSSION OF RESULTS

The total cross sections for the inelastic processes are obtained by integrating Eq. (19) with respect to the impact parameter ρ. Since the phases χ_1 and χ_2 are very dependent on the impact parameter, the terms containing $\cos(\chi + \chi_1)$, $\cos(\chi + \chi_2)$, and $\cos(\chi - \chi_2)$ in Eq. (22) do not contribute to the total cross sections. The term containing $\cos(\chi^+ + \chi_1 - \chi_2)$[1] will also be ignored. Therefore, we retain only the last term in Eq. (22).

To obtain explicit expressions for the total cross sections, let us consider the case where γ_1 and γ_2 are small. This may occur, for example, because the ground-state term of the system in the region of R_1 and R_2 varies much more rapidly than terms 1 and 2. The oscillating part of the total cross section is then

$$\Delta Q = 8\pi^2[p_3(1-p_3)]^{1/2}\int_0^{\rho_{max}}\sqrt{\gamma_1\gamma_2}\cos\chi(\rho)\,\rho\,d\rho. \qquad (24)$$

Let us consider the behavior of χ in more detail. For small γ_1 and γ_2 we have $\varphi_1 = \varphi_2 = \pi/4$. The second integral in Eq. (23) is much less than the first, since $E_2 - E_0$ does not exceed $E_2 - E_1$ in the interval (t_2, t_1) and the interval $t_1 - t_2$ is itself much less than $t_3 - t_1$. To transform the first integral in Eq. (23), let us replace t by the nuclear separation $R = (\rho^2 + v^2t^2)^{1/2}$. We then have

[1] The difference $\chi_1 - \chi_2$ may turn out to be small, if terms 1 and 2 are close for $R < R_2$ as well. The term including $\cos(\chi + \chi_1 - \chi_2)$ must then be taken into account and must be combined with the last term in Eq. (22).

$$\chi(\rho)=\frac{1}{\hbar v}\int_{R_1}^{R_3}[E_2(R)-E_1(R)]\left[1-\frac{\rho^2}{R^2}\right]^{-1/2}dR+\varphi_3. \quad (25)$$

If $\cos\chi(\rho)$ undergoes a large number of oscillations as ρ varies from 0 to R_2, then ΔQ will be small, and the oscillation structure of the total cross sections will not appear. To ensure that integration with respect to the impact parameter does not lead to the appearance of oscillations, it is necessary for the phase $\chi(\rho)$ to be a slow function of ρ. We recall that φ_3 is independent of ρ.

Let us write

$$\chi(\rho)=[u_0+u(\rho)]/v+\varphi_3, \quad (26)$$

where

$$u_0=\frac{1}{\hbar}\int_{R_1}^{R_3}\Delta E(R)dR, \quad (27)$$

$$u(\rho)=\frac{1}{\hbar}\int_{R_1}^{R_3}\Delta E(R)\left[\left(1-\frac{\rho^2}{R^2}\right)^{-1/2}-1\right]dR, \quad (28)$$

$$\Delta E(R)=E_2(R)-E_1(R). \quad (29)$$

Assuming that $R_3 \gg R_1$ and that $\rho < R_1$, we can use Eqs. (27) and (28) to show that

$$u_0 \approx R_3\Delta E/\hbar, \quad u(\rho) \approx R_1\Delta E'/\hbar. \quad (30)$$

In these expressions, ΔE and $\Delta E'$ are certain mean values of the difference $E_2 - E_1$, where for $\Delta E'$ the important region of distances is of the order of R_1. For $R_3 \gg R_1$ we can find an interval of the velocity v in which

$$R_1\Delta E'/\hbar < v < R_3\Delta E/\hbar. \quad (31)$$

In this velocity interval we can neglect $u(\rho)/v$ inside the cosine so that $\cos\chi$ can be taken from under the integral sign in Eq. (24). So far, we have assumed that the colliding particles move uniformly along straight lines. This restriction is, however, unimportant. In the region of threshold energies, integrals of the form $\int(E_a - E_b)dt$ were replaced by action differences $S_b - S_a$. Equation (24) is not affected by this because it has already been averaged over the oscillations connected with double transits of the crossing points R_1 and R_2.

After integration with respect to ρ in Eq. (24), we finally obtain

$$\Delta Q=[Q_{1L}Q_{2L}p_3(1-p_3)]^{1/2}\cos\chi(0), \quad (32)$$

where

$$Q_{kL}=8\pi^2V_k^2R_k^2\left(v^2-\frac{2E_k}{\mu}\right)^{1/2}\Big/\hbar|\Delta F_k|v^2 \quad (33)$$

is the Landau cross section,[13] μ is the reduced mass of the colliding particles, and the quantities V_k^2, E_k, and $|\Delta F_k|$ are taken at the pseudocrossing point $R = R_k$.

The total cross sections for the inelastic processes in this approximation are given by

$$Q_1=Q_{1L}p_3+Q_{2L}(1-p_3)+\Delta Q, \quad (34)$$

$$Q_2=Q_{1L}(1-p_3)+Q_{2L}p_3-\Delta Q. \quad (35)$$

In Eqs. (32), (34), and (35) we have neglected the difference between R_1 and R_2 and between the energies of the terms E_1 and E_2 at the crossing points. It is not difficult to take these differences into account but the final formulas are much more complicated. Equations (32)–(35) are valid at energies exceeding the threshold energy by an amount greater than $E_2 - E_1$ at the crossing points.

We recall that $p_3 = e^{-2\pi\gamma_3}$ in Eqs. (32), (34), and (35) if the term interaction during the separation of the particles occurs as a result of crossing. Here we have $\gamma_3 = V_3^2/\hbar v|\Delta F_3|$. On the other hand, if the interaction during the particle separation is connected with term approach, then $p_3 = (1 + e^{-\pi\delta})^{-1}$, where δ is given by Eq. (17).

It is thus clear from the final formulas given by Eqs. (32), (34), and (35) that, in the velocity interval defined by (31), the interference of the two states during the separation of the particles leads to oscillations in the total cross sections for the inelastic processes defined by Eq. (1).

Since the contribution to the oscillating part of the total cross section gives only the last term in Eq. (22), which is due to interference between paths $0A'B'B_0B2$ and $0A'B'B_0BA1$ in Fig. 2 (so that the interfering paths coincide before B), the phase of the oscillations in the total cross sections is independent of the behavior of the terms for $R < R_2$. Therefore, if in this region there are term interactions which are not taken into account in this model, this will not change the phase of the oscillations although their amplitude may be modified.

Let us now consider the oscillation phase as a function of velocity. In the case of term approach, the phase of the oscillations is

$$\chi(0)=\frac{1}{\hbar v}\int_{R_1}^{R_3}\Delta E(R)dR+\frac{H_{22}-H_{11}}{\hbar\lambda v}. \quad (36)$$

It is clear that, in this case, the total cross sections are harmonic functions of the reciprocal of the velocity.

In the case of term crossing, the quantity φ_3 which is present in the definition of $\chi(0)$ is a complicated function of velocity (see Fig. 12). The oscillations can then be harmonic as functions of $1/v$ in two limiting cases, namely: if $\gamma_3 \ll 1$ then $\varphi_3 = \pi/4$, and

$$\chi(0)=\frac{1}{\hbar v}\int_{R_1}^{R_3}\Delta E(R)dR+\frac{\pi}{4}; \quad (37)$$

while for $\gamma_3 > 1$ we have $\varphi_3 = \gamma_3$, and

$$\chi(0)=\frac{1}{\hbar v}\int_{R_1}^{R_3}\Delta E(R)dR+\frac{V_3^2}{\hbar v|\Delta F_3|}. \quad (38)$$

We note that if γ_3 or δ is large in comparison with unity, then p_3 is small, and this means that term interaction during the separation of the particles does not, in fact, occur. Consequently, if the experimental modulation depth is not small [according to Eqs. (32), (34), and (35), the maximum modulation depth is 50%], we may conclude that the second terms in Eqs. (36) and (38) are small, and the period of the oscillations on the reciprocal velocity scale is practically equal to $2\pi/u_0$, where u_0 is given by Eq. (27) and is determined by the area between the interacting terms in the interval between the population point R_1 and the interaction point R_3 (shown shaded in Fig. 2).

When the experimental energy dependence of the total inelastic cross section is analyzed, the following conclusions, drawn from our calculations, are useful. If the experimental curve exhibits a large number of oscillation periods, one would expect the extreme to be equally spaced (on the 1/v scale), at least for high velocities.

If the reason for the term interaction during particle separation is the term approach, the modulation depth at high velocities should be velocity-independent (if $\delta \to 0$, $p_3 \to 0$). If, on the other hand, term crossing occurs during the separation of the particles, the modulation depth may decrease at high velocities ($p_3 \to 1$ if $\gamma_3 \to 0$). At low velocities, the oscillations in the total cross section may become irregular and not so well defined after averaging over the impact parameter [violation of (31)] and as a result of the reduction in the effectiveness of the interaction with decreasing velocity ($p_3 \to 0$ when $\gamma_3 \to \infty$ and $p_3 \to 1$ when $\delta \to \infty$; in both cases the mixing of states disappears).

Equations (32), (34), and (35) were obtained on the assumption that γ_1 and γ_2, describing the population of terms 1 and 2, were small. For arbitrary γ_1 and γ_2 the corresponding expressions are rather complicated. There is, however, an interesting qualitative effect which may occur in this case. The coefficient of $\cos\chi$ in Eq. (22) contains the factor $(1-2p_2)$ which vanishes when $p_2 = 1/2$. Therefore, in the neighborhood of the corresponding energy, the regularity of the oscillations may be violated, and the phase may change by π.

It is important to note that, in this paper, we have considered only a simple model which leads to oscillations in the total cross sections for two inelastic channels.

The appearance of oscillations in the excitation probability for a given state of a set of two atomic particles is, in fact, the result of the existence of a number of alternative paths leading to this state (this also refers to Landau-Stückelberg oscillations). To each pair of paths there corresponds an oscillation mode whose phase is determined by the area ($\int \Delta E dt$) bounded by these two patches on the diagram showing the energy of the system as a function of time. If this area is a slowly varying function of the impact parameter, then the oscillations will appear in the total cross sections as well, and the frequency of these oscillations on the 1/v scale will be proportional to the area bounded by these two paths on the diagram showing the energy of the system as a function of the nuclear separation. Consider the case shown in Fig. 4, where three terms interact at large nuclear separations. This situation differs from that shown in Fig. 4 only by the fact that the coherent population of states 1 and 2 is achieved not directly from the ground state 0 but through the term 3. Oscillations in the probability of population of states 1 and 2 are, in this case, connected with the presence of the closed loop ABC. If the behavior of the quasimolecular terms at large separations leads to the formation of such a loop, the probability oscillations are repeated in the total cross sections for the inelastic channels 1 and 2 because the area of the loop is practically independent of the impact parameter. The final formulas for the cross sections are then similar to those obtained above.

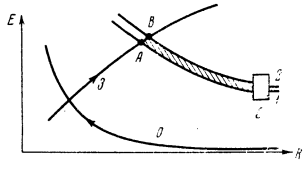

FIG. 4

If a number of inelastic channels interact at large nuclear separations, there can be a number of groups and, correspondingly, a number of oscillation modes on the cross sections for these channels. These cases can be treated by a simple application of Eq. (3) at each point of the nonadiabatic term interaction.[2)]

We note that one would expect to see the oscillations on the total cross sections not only when the term interaction leading to the formation of a closed loop occurs during the separation of the particles, but also when this interaction occurs during their approach. An example of this is afforded by transitions from states 2 or 1 to states 0 in Figs. 1 and 4.

In conclusion, let us consider some experimental data. There are a few examples in the literature of the oscillation structure of the total inelastic cross sections for processes with large resonance defects in slow collisions of heavy atomic particles.[5-8,14,15]

Very regular oscillations were found in[7] for the process

$$Na^+ + Ne \to Na^+ + Ne^* - 16{,}85\ eV \qquad (39)$$

Figure 5a shows the intensity of the λ 736 Å line of Ne I corresponding to the excitation of the resonance level $2p^5(^2P^0_{1/2})3s^1P^0_1$ as a function of the reciprocal velocity of relative motion of the colliding particles. The points in Fig. 5b show the experimental values of $\Delta Q/Q_m$, where Q_m is the smooth part of the measured cross section and ΔQ is the oscillating part. The solid curve is the graph of the function $0.25\cos(u_0/v + \pi/4)$ for $u_0 = 2.30 \times 10^8$ cm/sec. The parameters of this

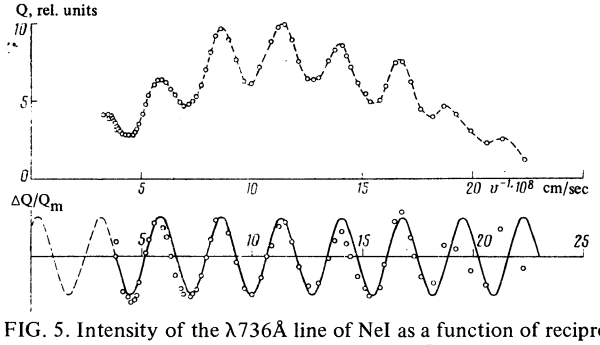

FIG. 5. Intensity of the λ736Å line of NeI as a function of reciprocal velocity for the process $Na^+ Ne \to Na^+ + Ne^*$ [7] (upper curve) and the oscillating part of this line intensity referred to the smoothly varying component (lower curve). Points are experimental, solid line represents the function $0.25\cos(2.3 \times 10^8/v + \pi/4)$; the broken line represents the part of the lower curve corresponds to high velocities for which experimental data are not available.

[2)]Three-channel interaction was considered in [16] in connection with oscillations on the charge-transfer cross section for Na^+ in neon.

function are chosen so that it provides the best fit to the observed oscillations in the velocity region where they are most regular. It is very clear from this figure that the oscillations are, in fact, regular. The modulation depth is quite high (25%) and is not very sensitive to the velocity. This shows that the population probabilities for the interacting levels are not very different, and the interaction during particle separation is sufficiently effective. It is clear from Fig. 5b that the phase of the oscillations tends to $\pi/4$ as $v \to \infty$, so that it would appear that, in this case, the term interaction at large distances is connected with term crossing [see Eq. (37)]. The quantity u_0 taken from experiment enables us to relate R_3 to the mean energy difference ΔE between the interacting levels. According to Eq. (27), $u_0 \approx R_3 \Delta E/\hbar$ and, if we suppose that $\Delta E \approx 1$ eV, then $R_3 \approx 15$ Å.

It is suggested in[7] that the second inelastic channel for the reaction (39) is the charge-transfer process

$$Na^+ + Ne \to Na^0 + Ne^+ - 16.42 \text{ eV} \qquad (40)$$

and that, therefore, the charge-transfer cross section should exhibit oscillations in antiphase with the oscillations in the total cross section for the excitation process defined by Eq. (39). Latypov and Shaparenko[16] have recently measured the charge-transfer cross sections for Eq. (40) and have shown that it does, in fact, exhibit these oscillations. However, the oscillation period is smaller by a factor of two as compared with the reaction (39). They suggest that this change in the period is associated with the influence of the third channel.

The excitation function for the λ 584.3 Å resonance line of He is reported in[17] for the process

$$Na^+ + He \to Na^+ + He^* - 21.22 \text{ eV} \qquad (41)$$

These results are shown in Fig. 6 on the 1/v scale together with the excitation cross section for the yellow sodium doublet Na($3p^2P_{1/2,3/2}$) during the process

$$Na^+ + He \to Na^* + He^+ - 21.55 \text{ eV} \qquad (42)$$

which is taken from the early paper by Maurer[18] after conversion to the 1/v scale. It is clear that the two cross sections oscillate in antiphase. The three maxima on the curve for reaction (41) are approximately equally spaced.

We are indebted to V. M. Dukel'skii for constant interest in this work, and to Yu. N. Demkov for useful discussions.

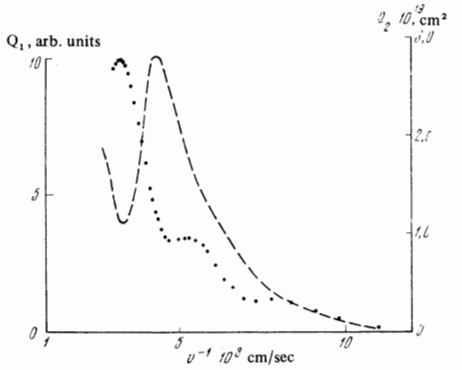

FIG. 6. Excitation cross section Q_1 for the λ584.3Å line of He for the process $Na^+ + He \to Na^+ + He^*$ [17] (shown by points), and the Mauer data [18] for the excitation cross section Q_2 of the yellow doublet of sodium in the reaction $Na^+ + He \to Na^* E He^+$ plotted against the reciprocal velocity and shown by the broken line.

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