Unified formula for the inelastic differential scattering cross sections of atoms and determination of the excited-term parameters

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The two-term crossing model is used to deduce a unified expression for the inelastic differential cross section which is valid throughout the angular range. It is shown that this expression correctly combines the formulas which were obtained previously and respectively describe the threshold rainbow behavior of the cross section and the quasiclassical interference at nonthreshold angles which is connected with the presence of two trajectories. A procedure is evaluated for the determination of the excited-term parameters using experimental inelastic data and the unified formula. The advantages and improvements which this formula gives as compared with previous expressions are discussed. Specific calculations are reported for $He^+ - Ne$ at a collision energy of 71 eV.

1. The oscillatory structure of the inelastic differential cross section for collisions between atoms, and its interpretation within the framework of the close-coupled two-state methods, are an important source of information for the determination of the quasimolecular terms responsible for the observed excitation.

An essential feature of the cross sections is the presence of rainbow effects at the threshold for elastic and inelastic processes. These are due to the minimum in the deflection function which occurs even for two strictly monotonic repelling terms, as was first pointed out in[1,2]. On the other hand, at angles well in excess of the threshold value, the rainbow oscillations should be replaced by the usual quasiclassical interference between the two trajectories which contribute to the inelastic process.

These two ranges of the scattering angle were considered in[2] within the framework of the high-energy approximation, and expressions were obtained for the inelastic cross section [see Eqs. (10) and (11)] which are valid, respectively, near the threshold angle and well away from this value. Previous attempts to determine the terms for $He^+ - Ne$, as reported in[2,3], were based on the use of some particular formula to represent the observed characteristic features of the oscillations (periods, size of the cross sections at first maximum, and so on) as functions of the collision energy.

The main defect of this approach is that the formulas which are used and the relationships which follow from them are of restricted validity insofar as the scattering angle is concerned, and do not pass one into the other although this is claimed in[2]. Thus, in the well-known series of papers by Smith et al., most of whose results are summarized in[3], the rainbow structure of the threshold behavior of the cross section[1)] is ignored altogether, and the characteristic excited-term parameters are deduced on the basis of the quasiclassical formula given by Eq. (11); at the same time, the necessity of deducing the measured threshold angle within the framework of their model leads Smith et al. to the parametrization of the excited potential, which includes attraction at distances close to the term-crossing point, and this is not necessary. On the other hand, in[2] the analysis of the first periods of the oscillations is based on Eq. (2), but it will not be shown below that this extension of the threshold dependence is not strictly valid because Eq. (10) is valid only within a very small range of angles near the threshold, and the description of even the first oscillation period requires a generalization of this formula.

The validity of any particular prediction insofar as the cross sections and the term parameters are concerned can, of course, be checked against specific calculations of cross sections, using the potential deduced from the analysis. Equations (10) and (11) are not suitable for a direct calculation of this kind for reasons indicated above. Our first aim in the present paper will therefore be to obtain a unified formula for the inelastic differential cross section which would be valid throughout the angular range and would ensure transition from the threshold behavior to the usual quasiclassical interference well away from the threshold. For the sake of simplicity, it is best to derive this formula in the high-energy approximation although similar results can be obtained in the general case.

2. If we consider transitions in a two-state system within the framework of the high-energy approximation, we can show that the reduced inelastic cross section $\rho(\tau) = \theta \sin\theta |f_{12}(\theta, E)|^2$ is given by[2]

$$\rho(\tau)=\frac{\tau}{\pi\hbar^3 v^3}\left|\int_0^\infty\int_{-\infty}^{\infty}\sqrt{b}\,V_{21}(R)\exp\left\{\frac{i}{\hbar}S(b,z,\tau)\right\}dz\,db\right|^2. \qquad (1)$$

where θ is the scattering angle, $f_{12}(\theta, E)$ is the scattering amplitude, E is the collision energy, $\tau = \theta E$ is the reduced angle, v is the relative velocity of the system, b is the impact parameter, R is the distance between the nuclei, $R^2 = b^2 + z^2$, and $V_{21}(R)$ is the matrix element for the interaction between the states. The action function is then given by

$$S(b,z,\tau)=\frac{2}{v}\left\{\int_0^\infty\frac{1}{R}\frac{dV_a(R)}{dR}z^2\,dz-\int_0^z V_d(R')dz'-b\tau\right\}, \qquad (2)$$

where

$$V_a(R)=\tfrac{1}{2}(V_1(R)+V_2(R)),\quad V_d(R)=\tfrac{1}{2}(V_1(R)-V_2(R)),$$

and $V_1(R)$ and $V_2(R)$ are the ground and excited terms of the system.

Integrating Eq. (1) by the stationary phase (SP) method with respect to z, we obtain

$$\rho(\tau)=\frac{2\pi|V_{12}(R_x)|^2R_x}{\hbar^2\Delta F v^2}|F(\tau)|^2, \qquad (3)$$

where $R(x)$ is the term-crossing point and $\Delta F = (dV_1/dR - dV_2/dR)|_{R_x}$ is the difference between the slopes of the potentials at the crossing point. The function $F(\tau)$ is given by

$$F(\tau)=\int_0^\infty \frac{\sqrt{b}}{\sqrt{z(b)}}\exp\left\{\frac{i}{\hbar}S(b,z(b),\tau)\right\}db. \quad (4)$$

The condition for the stationary phase with respect to z defines the form of the branches z(b) and, in particular,

$$z^2(b)+b^2=R_x^2. \quad (5)$$

The condition of stationary phase with respect to b for Eqs. (4) and (5) gives the well-known expression[2] for the branches of the deflection function $\tau_i(b)$, i = 1, 2 (see Fig. 1). Moreover, as expected, the two stationary phase points for Eq. (4) become identical in the neighborhood of the minimum (threshold) angle τ_{th}. In contrast to[2], instead of considering separately the case of different stationary points and the degenerate case, we shall use the device described in[4] for single-potential scattering when we integrate Eq. (4), and will replace the actual behavior of the stationary-phase points of Eq. (4) by the behavior of the stationary points of an Airy-type integral. This can be done by substituting

$$S(b,z(b),\tau)=a(\tau)x+{}^1/_3x^3+A(\tau). \quad (6)$$

where x is a new integration variable, and $a(\tau)$ and $A(\tau)$ are unknown transformation coefficients.

Substituting Eq. (6) in Eq. (4), we obtain

$$F(\tau)=\exp\left\{\frac{i}{\hbar}A(\tau)\right\}\int\frac{\sqrt{b(x)}}{\sqrt{z(x)}}\frac{db(x)}{dx}\exp\left\{\frac{i}{\hbar}\left(a(\tau)x+\frac{1}{3}x^3\right)\right\}dx. \quad (7)$$

The final expression for the cross section is obtained through the asymptotic expansion of Eq. (7), which is uniform in τ. To obtain the first term of this expansion the pre-exponential factor in Eq. (7) must be written in the form

$$\frac{\sqrt{b(x)}}{\sqrt{z(x)}}\frac{db(x)}{dx}=p+qx,$$

where p and q are unknown coefficients, and the mutually single-valued correspondence between the stationary phase points of Eqs. (4) and (7) must be taken into account so that the coefficients $a(\tau)$, $A(\tau)$, p, and q can be determined. The result of all this is a unified formula for the inelastic cross section which is valid for all $\tau \geq \tau_{th}$. This formula is

$$\rho(\tau)=\frac{|a(\tau)|^{1/2}}{\hbar^{1/2}}\left\{(\sqrt{\rho_1p_1}+\sqrt{\rho_2p_2})^2\Phi^2\left(-\frac{|a(\tau)|}{\hbar^{2/3}}\right)+\frac{\hbar^{2/3}}{|a(\tau)|}(\sqrt{\rho_1p_1}-\sqrt{\rho_2p_2})^2\Phi'^2\left(-\frac{|a(\tau)|}{\hbar^{2/3}}\right)\right\}. \quad (8)$$

where $\rho_i=\tau b_i(\tau)db_i(\tau)/d\tau$, i = 1, 2 is the classical expression for the cross section,

$$p_i=\frac{2\pi|V_{12}(R_x)|^2}{\hbar\Delta Fv\sqrt{1-b_i^2(\tau)/R_x^2}}$$

is the transition probability at the term crossing point, and $\Phi(x)$ and $\Phi'(x)$ are the Airy function and its derivative, respectively.

Using the above correspondence between the stationary-phase points in Eq. (6), we can readily show that

$$A(\tau)={}^1/_2(S_1(\tau)+(S_2(\tau)),\ a(\tau)=-|{}^3/_4(S_2(\tau)-S_1(\tau))|^{2/3}, \quad (9)$$

where $S_i(\tau)=S(b_i,z_i,\tau)$; b_i, z_i(i = 1, 2) are the stationary points of the integral given by Eq. (4).

It is readily seen that Eq. (8) does correctly combine the limiting formulas reported in[2]. We now use in the neighborhood of τ_{th} the expressions for $\Delta S(\tau)=S_2(\tau)-S_1(\tau)$ and $b(\tau)$, which correspond to the expansion of $\tau(b)$ to within terms of the second order in

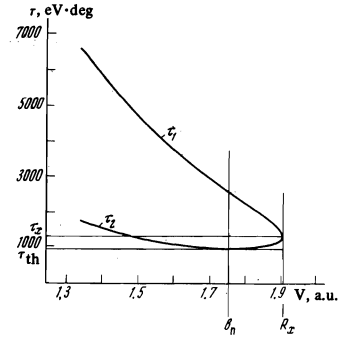

FIG. 1. Form of the branches $\tau_1(b)$ and $\tau_2(b)$ of the deflection function which contribute to the inelastic process in He^+-Ne collisions. τ_{th} is the threshold scattering angle, τ_x corresponds to the impact parameter $b = R_x$, where R_x is the term crossing point, and $b_{th} = b(\tau_{th})$.

$\Delta b = b - b_{th}$, and note that, in this region, the first term in Eq. (8) predominates over the second. This yields the expression for the cross section when $\tau \to \tau_{th}$:

$$\rho(\tau)=4\hbar^{-1/3}p_{th}\tau b_{th}\varkappa^{-2/3}v^{-1/3}\Phi^2(-2\hbar^{-2/3}v^{-2/3}\varkappa^{-1/3}(\tau-\tau_{th})),\ \tau\to\tau_{th}. \quad (10)$$

where $b_{th}=b(\tau_{th})$, $p_{th}=p(\tau_{th})$, $\varkappa=d^2\tau/db^2|b_{th}$.

In the other limiting case of large $\tau - \tau_{th}$, we can use the asymptotic expression for the Airy function and its derivative, and this yields

$$\rho(\tau)=\rho_1p_1+\rho_2p_2-2\sqrt{\rho_1p_1\rho_2p_2}\cos\left\{\frac{i}{\hbar}\Delta S(\tau)+\frac{\pi}{2}\right\},\quad \tau>\tau_{th}. \quad (11)$$

Thus, by analyzing Eq. (8) and its limiting cases, we find that the second term in this expression is important for its transition into Eq. (11), i.e., it is essential for the correct combination of Eqs. (10) and (11).[2)] We also note that Eq. (8) is valid for $\tau \geq \tau_{th}$. For $\tau < \tau_{th}$ (see Fig. 1), we can readily obtain the expression for the cross section but, in practice, the attenuation of the cross section for $\tau < \tau_{th}$ can adequately be described by Eq. (10).

Comparison of Eqs. (8), (10), and (11) shows the range of validity of Eqs. (10) and (11) and, in particular, it is readily demonstrated that the properties of the Airy function and its derivative ensure that, in fact, Eq. (8) does become identical with the quasiclassical interference given by Eq. (11), beginning with the second period of the oscillations.

The above approximate expression given by Eq. (8) can be directly verified by comparing existing quantum-mechanical calculations of the $He^+ - Ne$ cross sections[3] based on the numerical solution of two coupled equations with the potential deduced by analyzing the experimental data.

Figure 2 shows the results of this comparison. In this calculation we have used the potentials and the interaction matrix element given in[3]. It is clear that calculations based on Eq. (8) not only avoid the laborious procedure involved in the quantum-mechanical calculations, but give an adequate agreement with the latter. The discrepancy in the oscillation phase can be largely ascribed to the use of the high-energy approximation in deriving Eq. (8). This discrepancy can be readily shown to increase with increasing angle.

3. Let us now return to the determination of the term parameters. We shall start with Eq. (8), which enables us to calculate the cross sections throughout the angular interval in which we are interested. Specific calcu-

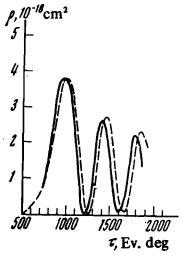

FIG. 2. Calculated cross sections using potentials taken from [3]. Broken curve—quantum-mechanical calculation from [3]. Solid curve—calculation based on Eq. (8) in the present paper.

TABLE I. Parameters for He^+-Ne

E, eV	Initial data			Calculated results						
	ΔV, eV.	A, at. un.	a, at. un.	c, at. un.	τ_{th}, eV deg	τ_x, eV.deg	R_x, eV.deg	b_{th} eV.deg	ΔF, eV.deg	$V_{21}(R_x)$, eV·deg
71	16,8	21,1	0,678	0,349	980	1335	1,91	1,76	—1,17	$8,6\cdot10^{-3}$

lations were carried out for the system He^+ – Ne, which was previously discussed in[2,3].

The use of experimental data on inelastic scattering enables us to introduce at least one adjustable parameter into the expression for the terms. For the He^+ – Ne pair we took the potentials in the form

$$V_1(R)=\frac{A}{R}e^{-R/a},\quad V_2(R)=\frac{A}{R}e^{-R/a}+\Delta V. \tag{12}$$

The parameters A and a were assumed known from the analysis of elastic data,[5] and ΔV is the known excitation energy of the system.

It follows that only one parameter, namely, c needs to be varied in the calculation. The other characteristic quantities, for example, R_X, b_{th}, τ_{th}, ΔF, and κ can readily be expressed in terms of c. This is achieved through the obvious result $V_1(R_X) = V_2(R_X)$ and the set of equations which define τ_{th} as a function of the deflection τ:

$$\tau(b_{th})=\tau_{th},\ d\tau/db|_{b_{th}}=0. \tag{13}$$

Essentially, the procedure is to fit the experimental data with the cross section calculated from Eq. (8).[3)] At the same time, we calculated the cross sections from Eqs. (10) and (11) in order to demonstrate the advantages of using Eq. (8). The results of calculations performed on the BESM-4 computer are shown in the table and in Figs. 1, 3, and 4.

It is clear from Fig. 3 that there is adequate agreement not only for the first periods of the oscillations are concerned, but also for the ratio of the amplitudes. This enables us to estimate $V_{21}(R_X)$ for He^+ – Ne, since it is clear from Eq. (8) that $|V_{21}(R_X)|^2$ is present in the expression for the cross section as a normalizing factor.

Comparison of theoretical predictions with the experimental data of Fig. 3, taken from our own work and from the similar curves given in Fig. 6 of[3], suggests that the parameters deduced in the present paper are, in fact, correct.

Comparison of calculations based, respectively, on Eqs. (8), (10), and (11), which is shown in Fig. 4, indicates that the unified formula gives a substantial improvement in the amplitude of the first period of the oscillations, and the transition to the quasiclassical asymptotic formula given by Eq. (11) occurs, as should be the case, during the second period of the oscillation

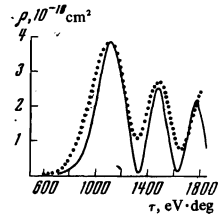

FIG. 3

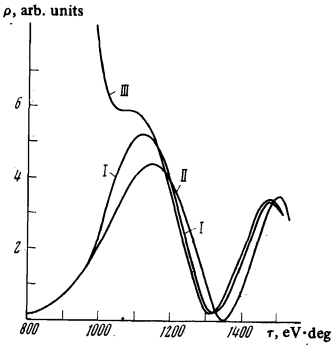

FIG. 4

FIG. 3. Inelastic differential cross section $\rho(\tau)$ for He^+-Ne; E = 71 eV, ΔV = 16.8 eV. Points—experimental data from [3]. Solid curve—calculations based on Eq. (8), using the potentials in the table.

FIG. 4. Calculated cross sections for He^+-Ne scattering, using the potentials defined in the table. I—calculation based on Eq. (8), II—calculation based on Eq. (10), III—calculation based on Eq. (11).

which, as noted above, restricts the range of validity of Eq. (10).

Two further points must be noted in connection with the above calculations. Firstly, the procedure used to determine the parameters of the excited term establishes only its relation to the main potential. Secondly, only one parameter is varied, and this is a relatively strong restriction. A substantial increase in the initial information can be achieved by considering analogous effects in the elastic channel (quasielastic oscillations), and this should lead to an increase in the number of adjustable parameters so that more accurate data can be obtained on interactions in the system of two crossing terms. It is clear that here again it would be useful to derive a unified formula for the description of the elastic cross section.

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1)We note that although the authors of [3] appreciate that the deflection function for the inelastic process has a minimum, they consider that scattering through angles less than τ_X (see Fig. 1) is connected with tunneling through a potential barrier (see p. 1614 of [3]).

2)Detailed arguments concerning the analogous case of single-potential rainbow scattering are reported in [4].

3)Since Eq. (8) is fitted to the experimental cross section in a restricted range of angles, it follows that the potential deduced in this way is, strictly speaking, limited on the side of small R by the smallest values of the impact parameter corresponding to scattering through the largest angle in this range, i.e., $R \gtrsim 1.3$ atomic units (Fig. 1).

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