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Origin of the left-right asymmetry in rotationally inelastic differential cross sections

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If the left ($\theta,\phi=0°$) and right ($\theta,\phi=180°$) rotationally inelastic differential cross sections are measured for the magnetic transitions, then in general the two results are not identical. It is shown, for a two-dimensional model and for linear molecules, that the origin of this asymmetry is due to multiple-collision effects of the atom with the molecule.

I. INTRODUCTION

In a typical atom-molecule collision experiment, with only rotational excitations, the differential cross section is measured for a particular transition $(0,0)\rightarrow(j,m)$ (for simplicity we assume that initially the molecule is not rotating) and at the given scattering angles (θ,ϕ). In most of the experiments the azimuthal angle ϕ is fixed while θ (the scattering angle) is variable, but one can also analyze the cross sections for a variable ϕ and fixed θ. Unlike a spherical target, when the cross section is ϕ independent, for a molecule, i.e., the nonspherical target, the cross sections will be ϕ dependent. In the ϕ dependence of the cross sections, two positions are of particular interest: $\phi=0°$ and $\phi=180°$, which is formally equivalent to the positions θ (right) and $-\theta$ (left) for the polar angles. In general, the two cross sections are not equal and the reason for this will be investigated in this work. We will find that the origin of this asymmetry has physical meaning, since it is due to multiple-collision effects of the atom with the molecule. To show this we will make use of the models for the rotational excitations. Let us therefore make a few comments on their use in the atom-molecule collisions.

Rotationally inelastic collisions of the atoms and molecules have been very thoroughly studied, both theoretically and experimentally.[1,2] However, when the theory is analyzed in more detail, we find two distinct approaches: one which deals with developing a suitable method for calculating accurate cross sections and the other which uses simple models to explain typical features of the cross sections. The latter approach is less accurate and not necessarily suitable for accurate comparison with experiment, but it has an advantage when we are interested in understanding the physics of the inelastic collision process. The former subject will not be of our immediate interest since nowadays there are powerful methods for computing the cross sections, although some further improvements must be taken into account.[3]

The use of simple models for explaining the features of the cross sections, in terms of some typical parameters of the potential surface, have not attracted great attention. Only recently this attempt was made using the hard-core ellipsoid model in connection with rotational rainbows.[4,5] The rotational rainbows were also studied using the Infinite-order sudden (IOS) limit of the Schrödinger equation[6,7] and when applied to hard-core ellipsoids, similar relationships were found as in the classical model.[8]

What is the physical justification for using such models? A typical atom-molecule potential consists of two parts: the tail and the intermediate region (where the well of the potential is included) and the highly repulsive core for small atom-molecule separation. Using order-of-magnitude estimates we will now show that, for the case of rotational excitations of molecules, the tail and the intermediate region do not give important contributions.

Any rotation is caused by a torque, and classically if this torque is applied in the time interval. Δt, the final rotational momentum of the molecule is

$$\hbar|j|=\Delta t\,|\vec{R}\times\vec{\nabla}V| \ , \tag{1.1}$$

where $\vec{R}$ is the vector from the center of mass of the molecule to the incoming atom, and $\vec{\nabla}V$ is the gradient of the potential. We immediately notice that at large distances the vector product in (1.1) is almost zero since the potential is almost spherical.

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At the intermediate distance, i.e., around the position of the minimum of the potential, $\vec{\nabla} V$ can have an angular component and we will assume that it is constant during the time Δt, in which case

$$\hbar j = \Delta t \left| \frac{\partial V}{\partial \phi} \right| , \tag{1.2}$$

where we have also assumed that the collision takes place in two dimensions. The expression (1.2) gives the maximum possible change in the rotational quanta j, in the intermediate range of R. If for Δt we give a typical value $\Delta t \sim 10^{-15}$ s (we should take into account that $\partial V/\partial \phi$ is large in a relatively small range of R) and $j=2$, i.e., the minimal change in the rotational quanta for the homonuclear molecule, then $\partial V/\partial \phi \sim 1$ eV/rad. This value is very large and only potentials with deep wells, e.g., ion-molecule potentials, can satisfy this condition. Otherwise this part of the potential indeed does not contribute very much to the inelastic transitions.

Therefore, only the short-range part of the potential, where the gradient is very large, will determine the essential structure of the cross sections. Of course, the details of the potential, i.e., the tail and the intermediate part, will contribute to the exact shape of the differential cross sections but will not alter the overall features such as the number of possible transitions, spacing of the broad oscillations in a differential cross section, and the positions of the peaks of the rotational rainbows. Therefore it is reasonable to replace the potential by a hard core, which is determined by the line where the kinetic energy of the system is zero. As has been argued,[5] the shape of the hard core, in many cases, is almost an ellipsoid, e.g., for the homonuclear molecules, but there are obvious cases where it is not.[9] However, it turns out that the results of this work, where we investigate the left-right symmetry of the cross sections, are also valid for a more general topology of the target. In fact, the proofs of Sec. III of this work can be repeated for a general shape of the target, but we have only shown it for the ellipsoid.

We will further simplify the ellipsoid model by assuming that the scattering occurs in two dimensions. The two-dimensional model greatly simplifies the mathematical part of the problem but at the same time does not neglect the basic structure of the scattering process. There are obvious limitations of the 2D models, but some properties of the differential cross sections are not altered by reducing the reducing the dimensionality. For example, the position of the rotational rainbows and the broad oscillations of the differential cross sections will remain unaltered when compared with the 3D calculations[10] and experiment.[11] Furthermore, the number of maximally accessible transitions is exactly predicted in 2D. The physical reason for this has already been discussed.[5] It was also recently shown that there is a propensity for preserving the j_z quantum number in 3D rotational collisions,[12] which is an indirect indication of the relevance of the 2D models.

From this short discussion we can safely assume that the 2D ellipsoid model has predictive power. Therefore, using this model, we will study in this paper one feature of the cross sections which will contribute to our understanding of rotationally inelastic collisions. One of the results of the study of the ellipsoid model in 2D is that the kinetic energy of the system can never be totally transferred into rotation if

$$\epsilon = \mu / I < 1/(A-B)^2 , \tag{1.3}$$

where μ is the reduced mass of the system and I is the momentum of inertia of the ellipsoid. The large and small axes of the ellipsoid are A and B, respectively. This conditions was derived under the assumption that initially the molecule was not rotating.

However, when $\epsilon = (A-B)^{-2}$ all the kinetic energy is transferred into rotation and the atom does not move away from the molecule. It is waiting to be hit by the other end of the rotating molecule. After this second collision the atom absorbs some of the rotational energy and flies away. The question is: What is the effect of this process on the cross sections? For brevity we will refer in the future to this effect as the MC (multiple collision) effect. Hence, how will the MC effect be observed in the differential cross section? In this paper we will show that the MC effect causes the left-right asymmetry of the differential cross section.

In Sec. II we will give more general conditions for the MC effect than (1.3), and in Sec. III we will discuss under what circumstances, both in classical and quantum mechanics, the left and right cross sections are symmetric. As will be shown, indeed the MC effect is the prime cause of the asymmetry.

II. CLASSICAL CONDITION FOR MC EFFECT

We have described in the Introduction the essential idea of the MC effect. However, the situation when the molecule absorbs all the kinetic energy is rather special. Let us therefore generalize the MC effect to all the processes when after the first collision the particle will have kinetic energy but may be scattered in such a direction that the ellipsoid will hit it a second time. The effect is not exactly the same as described in the Introduction but the chain of events is identical.

In the Introduction we have given the condition for the simple MC effect and here we will derive the condition for this more general effect. Let us first define a suitable coordinate system. In the center of the ellipsoid we place the coordinate system with its negative y axis pointing towards the incoming particle. The coordinates of the point where the particle hits the ellipsoid are

$$x=b\ ,\qquad y_b=[b(A^2-B^2)\sin\alpha\cos\alpha-AB(A^2\cos^2\alpha+B^2\sin^2\alpha-b^2)^{1/2}]/(A^2\cos^2\alpha+B^2\sin^2\alpha)\ . \tag{2.1}$$

After collision the particle is scattered in the direction ω with respect to the x axis. The equation of motion is

$$x=b+\hbar\frac{k}{\mu}t\cos\omega\ ,\qquad y=y_b+\hbar\frac{k}{\mu}t\sin\omega\ , \tag{2.2}$$

where $\hbar$ is the reduced Planck constant and k is the wave number after collision, i.e., $k=(k_0^2-\epsilon j^2)^{1/2}$. It is assumed that the time $t=0$ is defined at the moment of impact.

The ellipsoid, after collision, is in the rotation state j. During time t it will rotate by an angle $\beta-\alpha$, given by

$$\beta-\alpha=\frac{j\hbar}{I}t\ . \tag{2.3}$$

If the value of t, obtained from (2.3), is replaced in (2.2) we obtain

$$x=b+\frac{[1-\epsilon(j^*)^2]^{1/2}}{j^*}(\beta-\alpha)\sin\theta\ ,\qquad y=y_b+\frac{[1-\epsilon(j^*)^2]^{1/2}}{j^*}(\beta-\alpha)\cos\theta\ , \tag{2.4}$$

where θ is the scattering angle, related to ω by $\omega=(\pi/2)-\theta$. We have also used the notation for the reduced rotation quantum number $j^*=j/k_0$, and henceforth we will assume that $j^*>0$. It should be pointed out that in two dimensions there are only two projections of the rotational angular momentum of the molecule on the z axis: $\pm j$. For simplicity, from now on we will assume the positive projection, but this restriction is not essential.

The necessary condition for the MC effect is that at the orientation β of the ellipsoid the coordinates (2.4) of the particle are equal to (2.1), where b is replaced by x of (2.4) and α by β. Figure 1 shows one such possibility. If we want to find the general condition for the MC effect in terms of the parameters of the ellipsoid, the set of equations of motion should be solved,[5] together with the conditions already mentioned. However, in general, this is not possible because the equations, although analytic, are difficult to solve. We can only derive an approximate condition which can be obtained from the following physical reasoning. For the impact parameter $b=A-B$, the angle $\cos(2\alpha_0)=(A-B)/(A+B)$, $\alpha_0<0$, and arbitrary ϵ, the incoming particle, is always scattered in the backward direction.[5] At the same time the ellipsoid will acquire the maximal possible rotation. Therefore, if the particle, while flying back, escapes the zone of the maximal reach of the ellipsoid before the tip of the ellipsoid comes to the particle, there will be no MC effect.

The point which can be maximally reached by the ellipsoid on the line of impact $x=b$ is when

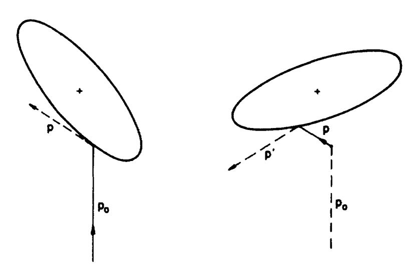

FIG. 1. MC effect. After the first impact (a) the atom is scattered in direction $\theta>180°$ with momentum p. The second impact with the ellipsoid (b) changes direction of the atom once again.

the end of the large axis is exactly on this line. The y value of this point is

$$y=-A\sin\gamma=-[B(2A-B)]^{1/2}\,, \tag{2.5}$$

where γ is defined as the angle between the axis A and the positive x axis. The angle γ is given by

$$\cos\gamma=(A-B)/A \tag{2.6}$$

and is related to β by

$$\beta=\pi-\gamma\,. \tag{2.7}$$

As we have mentioned, the condition for the MC effect is that (2.5) should equal (2.4), i.e.,

$$-[B(2A-B)]^{1/2}=-(AB)^{1/2}-\frac{[1-\epsilon(j^*)^2]^{1/2}}{\epsilon j^*}(\beta-\alpha_0)\,, \tag{2.8}$$

where we have taken into account that for the angle α_0, the point of impact (2.1) is equal to $y_b=-\sqrt{AB}$. Since the value of j^* in such a case is[5]

$$j^*=2(A-B)/[1+\epsilon(A-B)^2]\,, \tag{2.9}$$

we obtain the value of ϵ which satisfies (2.8)

$$\epsilon=\frac{(\beta-\alpha_0)}{\{d[2B(\sqrt{A+d}-\sqrt{A})+d(\beta-\alpha_0)]\}}\,, \tag{2.10}$$

where β is given by (2.6) and (2.7) and $d=A-B$. It can be shown that ϵ, given by (2.10), is always smaller than $\epsilon=(A-B)^2$, the condition derived in the Introduction. Therefore, for an ellipsoid with ϵ smaller than (2.10) there will be no MC effect while for ϵ greater than (2.10) we will observe the effect. In this respect (2.10) is the lower bound on ϵ which gives the MC effect. However, this limit is only approximate for two reasons: (a) Because of the curvature near the tip of the ellipsoid, the lowest point in the negative y direction on the line $x=b$ is not (2.5). As a result the bound (2.10) will be somewhat lower but not significantly so. (b) The most favorable condition for the MC effect is not when the particle is scattered in $\theta=180°$. For example, if the particle, after the first impact, is scattered in $\theta>180°$ it will go towards the ellipsoid (because of our assumption that j is positive), and the chance of the second impact will be greater than the chance when it is scattered in $\theta=180°$. On the other hand, if the particle is scattered in $\theta<180°$ the ellipsoid must catch up with the particle, in which case the ellipsoid will not be able to hit the particle the second time. Therefore, scattering in $\theta>180°$ gives more favorable conditions for the MC effect than scattering in $\theta=180°$ or $\theta<180°$. As the result, the bound (2.10) on ϵ will be lower, about 10–20 %.

We have given in (2.10) the lower bound on ϵ The upper bound can be obtained in a similar manner. Let us assume that for large ϵ there are parameters b and α for which all the energy of the incoming particle is transferred into the rotation of the ellipsoid. The spin of the ellipsoid will be in such a case $j^*=1/\sqrt{\epsilon}$. From the general expression for the final rotational state[5]

$$j^*=2b_n\cos(\phi)/(1+\epsilon b_n^2)\,, \tag{2.11}$$

where

$$b_n=b\cos\phi+y_b\sin\phi \tag{2.12}$$

and

$$\tan\phi=[(A^2-B^2)\sin\alpha\cos\alpha+ABb/(A^2\cos^2\alpha+B^2\sin^2\alpha-b^2)^{1/2}]/(A^2\cos^2\alpha+B^2\sin^2\alpha)\,, \tag{2.13}$$

where ϕ is the angle between the initial momentum of the particle and the normal to the ellipsoid at the point of impact, we find two solutions: $\phi=0$ and $b_n=1/\sqrt{\epsilon}$. From (2.12) it follows that $b=1/\sqrt{\epsilon}$ and from (2.13)

$$\alpha\sim-B/[A^2-B^2)\sqrt{\epsilon}]\,, \tag{2.14}$$

where we have assumed $\alpha\sim0$. Therefore, for large values of ϵ there are always conditions for the MC effect.

III. SYMMETRY OF DIFFERENTIAL CROSS SECTION

We have seen that the MC effect is essentially a double collision effect. Furthermore, we have seen that the greatest chance for this effect is when the particle, after the first impact, is scattered in $\theta>180°$. We have also found that below a certain value of ϵ, given by (2.10), there will be no such effect. There are several conclusions which can be deduced from these facts. (a) If the value of ϵ is below the critical value for the MC effect, the left and right differential cross sections are equal, but if the value of ϵ is greater than the left cross section will be smaller compared to the right cross section for large inelastic transitions. (b) The position of the first maxima in the differential cross section (the rotational rainbow) is the same for the left and right space[5] in the absence of the MC effect. If the effect is present, the left position of the maxima will be shifted towards larger angles (for a pictorial explanation see Fig. 1). (c) After the second impact the ellipsoid will lose part of the rotation energy; therefore in the left space the small transitions, including the elastic one, will have non-negligible contributions from the higher-order process such as the MC effect, in addition to the direct process. The last effect is difficult to check but the first two can be, in principle, verified in an experiment. However, we still have to prove that the MC effect is the cause of the left-right asymmetry of the differential cross section. Our conclusion was based on the assumption that in the absence of the MC effect the left-right cross sections are identical. Therefore we must prove that indeed this is true.

One particular situation when there is no MC effect is for $\epsilon=0$, i.e., in the static limit. In such a case the momentum of inertia of the ellipsoid is infinite and on impact the ellipsoid will not rotate (hence the name static limit), therefore there will be no chance for the MC effect. Let us prove that in such a limit the differential cross sections are symmetric with respect to change $\theta\to-\theta$, which is equivalent to going from the left in the right scattering space. We will prove this for a general potential which has an axial symmetry (i.e., for the linear molecules), and therefore the same will apply also for an ellipsoid.

The 2D equation for the scattering of an atom on a molecule is

$$\frac{\partial^2\psi}{\partial x^2}+\frac{\partial^2\psi}{\partial y^2}+\epsilon\frac{\partial^2\psi}{\partial\phi^2}=(V-k^2)\psi\ , \tag{3.1}$$

where V is a function of $\cos(\phi-\theta)$ (the assumption of the axial symmetry). If we replace ψ by an expansion

$$\psi=\frac{1}{\sqrt{r}}\sum_{J=-\infty}^{\infty}\sum_{j=-\infty}^{\infty}\varphi_j^J(r)e^{iJ\theta+ij(\phi-\theta)} \tag{3.2}$$

we obtain, in the static limit ($\epsilon=0$), a set of coupled equations for φ_j^J:

$$\frac{d^2\varphi_j^J}{dr^2}=\frac{(J-j)^2}{r^2}\varphi_j^J-k^2\varphi_j^J+\sum_{j'}V_{|j-j'|}\varphi_{j'}^J\ , \tag{3.3}$$

where $V_{|j-j'|}$ are independent of J. We can now define the Jost functions from the asymptotic limit of φ_j^J for large r.

$$\varphi_j^J=Y_{j,j'}^J(+1)e^{ikr}+Y_{j,j'}^J(-1)e^{-ikr}\ . \tag{3.4}$$

From the form of the equation (3.3) we can deduce this symmetry property of φ_j^J:

$$\varphi_{j,j'}^{J+\Delta}=\varphi_{j+\Delta,j'}^J\ , \tag{3.5}$$

where Δ is some integer. The Jost functions also have this symmetry, hence

$$Y_{j,j'}^{J+\Delta}=Y_{j+\Delta,j'}^J\ . \tag{3.6}$$

The S matrix, which is defined as

$$S_{j,j'}^J=\{\,Y(+1)[Y(-1)]^{-1}\,\}_{j,j'}^J\exp\left[i\frac{\pi}{2}(2J-j-j'+1)\right] \tag{3.7}$$

will therefore have the following symmetry:

$$S_{j,j'}^{J+\Delta}=e^{2i\pi\Delta}S_{j+\Delta,j'+\Delta}^J \tag{3.8}$$

which can be easily checked from (3.6) and (3.7). However, it should be emphasized that in the derivation of (3.8) we have implicitly assumed that the set of equations (3.3) are defined with the initial conditions at some $r=R\neq0$, for which $\varphi_j^J=0$.

Similarly, we can also prove another symmetry of the S matrix, which comes from the fact that the set (3.3) is symmetric to the simultaneous change $J\to-J$ and $j\to-j$. It follows that

$$S_{j,j'}^{-J}=S_{-j,-j'}^J\exp[-2iJ\pi-i(j+j')\pi]\ . \tag{3.9}$$

Let us now show the $\theta\to-\theta$ symmetry property of the scattering amplitude. The scattering amplitude in 2D is

$$F_{j,j'}(\theta)=\sum_{J=-\infty}^{\infty}(S_{j,j'}^J-\delta_{j,j'})e^{iJ\theta}\ . \tag{3.10}$$

For $j\neq j'$ we make the change $\theta\to-\theta$ in (3.10), hence

$$F_{j,j'}(-\theta)=\sum_J S_{j,j'}^J e^{-iJ\theta}=\sum_J S_{j,j'}^{-J}e^{iJ\theta}\ . \tag{3.11}$$

If we use (3.19) we obtain

$$F_{j,j'}(-\theta)=\sum_J S_{-j,-j'}^J\exp[iJ\theta-2iJ\pi-i(j+j')\pi]\ , \tag{3.12}$$

and after using (3.8) with $\Delta=-j-j'$,

$$F_{j,j'}(-\theta)=e^{i(j+j')\pi}\sum_J S_{j,j'}^{J-j-j'}e^{iJ\theta}\ , \tag{3.13}$$

or when the summation index is shifted:

$$F_{j,j'}(-\theta)=F_{j,j'}(\theta)\exp[i(j+j')\theta+i(j+j')\pi] \tag{3.14}$$

We have therefore proved that the left and right scattering amplitudes, in the static limit, are identical except for a phase which disappears in the differential cross section. The phase in (3.14) only indicates that the waves contributing to the left and right scattering space have a different flow. The symmetry (3.14) is valid for any potential which has an axial symmetry.

Indeed in the static limit the left and right cross sections are identical. When $\epsilon\neq0$ but small, the symmetry will be preserved simply because the cross section will not change appreciably.[13] However, for large ϵ, the symmetry will be broken if for no other reason than the MC effect. We have already hinted that this symmetry comes entirely because of this effect by proving the symmetry of the cross sections in the static limit. We would like to give now a better indication that this is indeed true. The hard-core ellipsoid model offers such a proof because we can distinguish two different processes: single and multiple collisions. In fact, the equations of motion for the ellipsoid model were obtained in a single impact case,[5] and the MC correction of this article determines the equations in the double collision. Therefore if we can prove that the equations of motion in a single collision case produce the symmetric cross section then all the asymmetry comes from the MC effect. To be more rigorous we will show this in the semiclassical limit.

After the first impact the particle is scattered in the angle[5]

$$\theta=\pi-(\phi+\phi_r)\ , \tag{3.15}$$

where ϕ is the angle (2.13) and ϕ_r, the recoil angle, is

$$\tan\phi_r=\frac{1+\epsilon b_n^2}{1-\epsilon b_n^2}\tan\phi\ . \tag{3.16}$$

The final rotational angular momentum of the ellipsoid is

$$j^*=\frac{2b_n\cos\phi}{1+\epsilon b_n^2}\ , \tag{3.17}$$

where it was assumed that initially the ellipsoid is nonrotating.

The scattering angle θ and j^* are functions of the impact parameter b and the orientation angle of the ellipsoid α. Therefore, if we fix the final j^*, then for a given b we can find α so that (3.17) can be satisfied. Within this value of α we obtain the scattering angle θ for a given b. The functional relationship $\theta^{j^*}(b)$ which we obtain is analogous to the deflection function in elastic collisions, but here it has an additional index corresponding to the chosen transition. In fact, there are two deflection functions[5] leading from two different impact parameters to the same θ for a given j^*. Hence the scattering amplitude is given as a sum[14,15]

$$F_{0,j}(\theta)=N_1^{-1/2}e^{i\delta_1}+N_2^{-1/2}e^{i\delta_2}\ , \tag{3.18}$$

where N_1 and N_2 are two Jacobians, given by

$$N_i=\mathrm{Det}\begin{vmatrix}\dfrac{\partial\theta}{\partial b_i}, & \dfrac{\partial j}{\partial b_i}\\[2mm] \dfrac{\partial\theta}{\partial\alpha_2}, & \dfrac{\partial j}{\partial\alpha_i}\end{vmatrix}\ ,\quad i=1,2 \tag{3.19}$$

and δ_1 and δ_2 are the phase shifts corresponding to these two trajectories. However, the absolute values of δ_1 and δ_2 are not essential for the differential cross section, only their difference. Therefore, to prove the symmetry of the cross sections under the reflection $\theta\to-\theta$, obtained from the equations (3.15)–(3.17), we must show that

$$|\delta_1^{(\theta)}-\delta_2^{(\theta)}|=|\delta_1^{(-\theta)}-\delta_2^{(-\theta)}| \tag{3.20}$$

and

$$N_i^{(\theta)}=N_i^{(-\theta)}\ , \tag{3.21}$$

where we have introduced the index to indicate formally that the two expressions (3.20) and (3.21) are evaluated for the left and right scattering angles.

Since the particle is freely moving before and after the collision, the phase difference (3.20) is equivalent to the requirement that the "optical path" difference for the left and right scattering should be the same. If we designate the two impact parameters which lead to the scattering angle θ by b_1 and b_2, and the appropriate y coordinate of the impact with the ellipsoid [given by (2.1)] by y_1 and y_2, respectively, then it follows that $|\delta_1-\delta_2|$ is a function of $|b_1-b_2|$ and $|y_1-y_2|$. If we replace θ by $-\theta$ then for the equality (3.20) to hold it is sufficient that

$$|b_1^{(\theta)}-b_2^{(\theta)}|=|b_1^{(-\theta)}-b_2^{(-\theta)}| \tag{3.22}$$

and

$$|y_1^{(\theta)}-y_2^{(\theta)}|=|y_1^{(-\theta)}-y_2^{(-\theta)}|\ . \tag{3.23}$$

Therefore we have to prove (3.21)–(3.23) in order to establish the symmetry

$$\sigma_{0,j}(\theta)=\sigma_{0,j}(-\theta)\ , \tag{3.24}$$

and hence the role of the MC effect in the left-right asymmetry of the cross sections.

Let us therefore look at some symmetry properties of the equations of motion. From (3.15) we find that $\theta\rightarrow-\theta$ is equivalent to

$$\phi+\phi_r\rightarrow-(\phi+\phi_r)\ , \tag{3.25}$$

and by noting that ϕ and ϕ_r are of the same sign [see Eq. (3.16)] we can assume that

$$\phi\rightarrow-\phi\ . \tag{3.26}$$

As will become obvious from further discussion, (3.26) is the necessary and sufficient condition for the transformation $\theta\rightarrow-\theta$.

Since j^* is the same for both θ and $-\theta$ by our assumption, then it follows from (3.17) that

$$b_b^{(\theta)}=b_n^{(-\theta)}\ . \tag{3.27}$$

From the definition of b_n (the shortest distance between the line perpendicular to the surface of the ellipsoid at the point of impact and the center of mass of the ellipsoid), it follows from (3.27) that the two points of impact from which the particle is scattered in the angles θ and $-\theta$ are identical. Therefore it is trivial to show that in such a case

$$|y_1^{(\theta)}-y_2^{(\theta)}|=|y_1^{(-\theta)}-y_2^{(-\theta)}| \tag{3.28}$$

and

$$|b_1^{(\theta)}-b_2^{(\theta)}|=|b_1^{(-\theta)}-b_2^{(-\theta)}|\ , \tag{3.29}$$

which are exactly the requirements (3.22) and (3.23).

Since the same point on the ellipsoid scatters the particle into the angles θ and $-\theta$, the two orientations of the ellipsoid are related through the equation

$$\alpha^{(-\theta)}=\alpha^{(\theta)}-2\phi\ , \tag{3.30}$$

while the two impact parameters are related by

$$b^{(-\theta)}=b^{(\theta)}\cos2\phi+y^{(\theta)}\sin2\phi\ . \tag{3.31}$$

With these results we can proceed to prove the remaining requirement (3.21) for the symmetry of the differential cross sections. We will have to evaluate the appropriate derivatives of θ and j with respect to b and α. Let us first observe [from (3.16) and (3.17)] that both θ and j are explicit functions of ϕ and b_n. Therefore we can write

$$\frac{\partial\theta}{\partial b}=-\frac{\partial\phi}{\partial b}-\frac{\partial\phi_r}{\partial b}=-\frac{\partial\phi}{\partial b}-\frac{\partial\phi_r}{\partial b_n}\frac{\partial b_n}{\partial b}-\frac{\partial\phi_r}{\partial\phi}\frac{\partial\phi}{\partial b}\ . \tag{3.32}$$

Similarly we can write all other derivatives. In such a case the Jacobian (3.19) is

$$N=\left[\frac{\partial\phi}{\partial\alpha}\frac{\partial b_n}{\partial b}-\frac{\partial\phi}{\partial b}\frac{\partial b_n}{\partial\alpha}\right]\times\left[\frac{\partial\phi_r}{\partial\phi}\frac{\partial j^*}{\partial b_n}-\frac{\partial\phi_r}{\partial b_n}\frac{\partial j^*}{\partial\phi}+\frac{\partial j^*}{\partial b_n}\right]. \tag{3.33}$$

The second factor can be evaluated explicitly, using the definitions (3.16) and (3.17). We find

$$\frac{\partial\phi_r}{\partial\phi}\frac{\partial j^*}{\partial b_n}-\frac{\partial\phi_r}{\partial b_n}\frac{\partial j^*}{\partial\phi}+\frac{\partial j^*}{\partial b_n}=4\cos(\phi)(1+\epsilon b_n^2)^{-2}\ . \tag{3.34}$$

However, the first parenthetical expression in (3.33) is more difficult to evaluate. Let us first use (2.12) to calculate the derivatives $\partial b_n/\partial b$ and $\partial b_n/\partial\alpha$. We find

$$\frac{\partial\phi}{\partial\alpha}\frac{\partial b_n}{\partial b}-\frac{\partial\phi}{\partial b}\frac{\partial b_n}{\partial\alpha}=\frac{\partial}{\partial b}\left[\frac{\partial y}{\partial\alpha}\cos\phi\right]. \tag{3.35}$$

In the next step we calculate $\partial y/\partial\alpha$ from (2.1). After some lengthly algebra it can be shown that

$$\partial y/\partial\alpha\cos\phi=b_n\ . \tag{3.36}$$

Therefore the Jacobian is

$$N=\frac{4\cos\phi}{(1+\epsilon b_n^2)^2}\frac{\partial b_n}{\partial b}\ . \tag{3.37}$$

We can now use the definition (2.12) and (2.13) to obtain the final form of the Jacobian

$$N=\left[1-\frac{A^2B^2\cos^4\phi}{(A^2\cos^2\alpha+B^2\sin^2\alpha-b^2)^2}\right]\frac{4}{(1+\epsilon b_n^2)^2}\ . \tag{3.38}$$

Although (3.38) is given in a closed form it is difficult to prove its symmetry with respect to $\theta\rightarrow-\theta$. It is much easier to do this for (3.37). If we rotate the point on the ellipsoid (b,y), together with the whole ellipsoid, by an angle $-\alpha$, the value of b_n will not change (which is obvious from the definition of b_n given earlier). In such a case the large axis of the ellipsoid will coincide with the x axis, and b_n will be given by

$$b_n=\frac{b_0(A^2-B^2)(A^2-b_0^2)^{1/2}}{A[A^4-b_0^2(A^2-B^2)]^{1/2}}\ , \tag{3.39}$$

where b_0 is the impact parameters of the new position of the rotated point (b,y). It is easy to show that b_0 is given by

$$b_0=b\cos\alpha+y\sin\alpha\ . \tag{3.40}$$

Therefore the derivative in (3.37) is

$$\partial b_n/\partial b=\partial b_n/\partial b_0\partial b_0/\partial b=\frac{\cos(\alpha-\phi)}{\cos\phi}\frac{\partial b_n}{\partial b_0}\ . \tag{3.41}$$

We can now look at the transformation property of N under the reflection $\theta\to-\theta$. In such a case $\phi\to-\phi$ and $b_n^{(\theta)}=b_n^{(-\theta)}$, and we obtain

$$N^{(-\theta)}=\frac{4\cos(\alpha^{(-\theta)}+\phi)}{[1+(b_n^{(-\theta)})^2]^2}\left|\frac{\partial b_n}{\partial b_0}\right|^{(-\theta)}\ . \tag{3.42}$$

The derivative $\partial b_n/\partial b_0$ is independent of α and taking into account (3.30) we indeed obtain

$$N^{(-\theta)}=N^{(\theta)} \tag{3.43}$$

and also the symmetry of the differential cross sections under the reflection $\theta\to-\theta$, since we have proved (3.20) earlier.

The result obtained is more general than only the proof of the symmetry for the classical cross sections, since we also take into account the quantum effects. Therefore we conclude that the only explanation of the asymmetry in the differential cross sections arises from the MC effect. As will be shown in Sec. IV, the exact quantum cross sections are in a very good agreement with the predictions.

IV. SEVERAL EXAMPLES

We will now give a few illustrative examples to verify our discussion. Since the onset of the MC effect occurs when the ratio $\epsilon=\mu/I$ satisfies certain conditions, as discussed in Sec. II, we will make a model calculation for a fixed A and B and variable ϵ. We take $A=2.26$ Å and $B=1.875$ Å, which are the parameters for Li-H_2 at $E=0.4$ eV. These values were obtained from the potential surface calculated for the same system.[16] The energy E in units of Å^{-2} is therefore $k^2=299.37$ Å^{-2}. The value of ϵ for the same system if $\epsilon=5.659$ Å^{-2}.

From the formula (2.10), and the given parameters of the ellipsoid, we can evaluate ϵ_0 for which

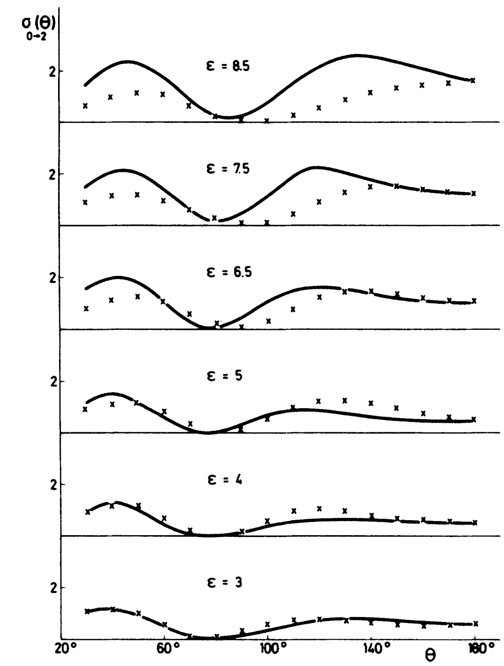

FIG. 2. Atom-ellipsoid differential cross sections for transition $j=0\to j=2$. The large axis of the ellipsoid is $A=2.26$ Å and the small axis is $B=1.875$ Å. The rotational constant ϵ is variable. The onset of the MC effect is for $\epsilon\sim4.3$ Å^{-2}.

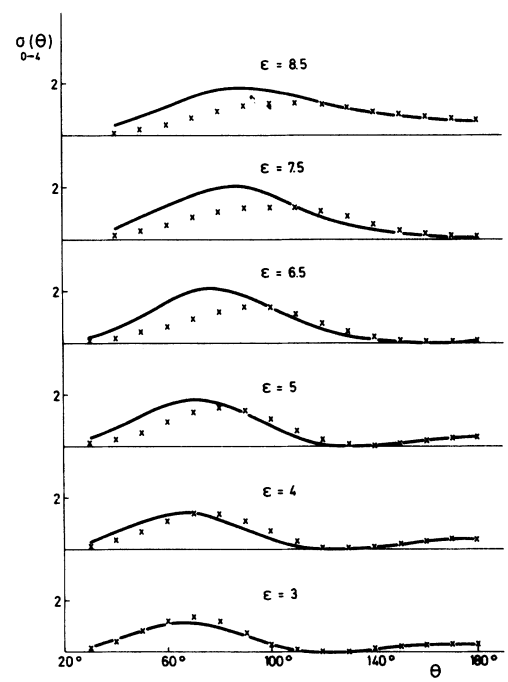

FIG. 3. Atom-ellipsoid differential cross sections for transition $j=0\to j=4$. The parameters are the same as in Fig. 2.

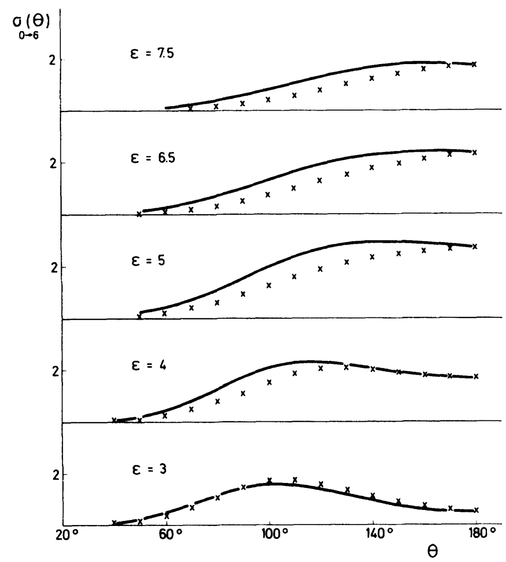

FIG. 4. Atom-ellipsoid differential cross sections for transition $j=0\rightarrow j=6$. The parameters are the same as in Fig. 2.

we have the onset of the MC effect. This value is $\epsilon_0=4.99$ Å^{-2}, which is well below ϵ for Li-H_2. ϵ_0 is even lower, for the reasons discussed in Sec. II, about $\epsilon_0\sim 4.3$ Å^{-2}. Therefore we expect that the MC effect will be observed, i.e., the left inelastic differential cross sections will not be equal to the right ones. In Figs. 2, 3, and 4 we give the inelastic differential cross section $0\rightarrow 2$, $0\rightarrow 4$, and $0\rightarrow 6$ (the elastic cross section is always symmetric) for a range of ϵ, from $\epsilon=3$ Å^{-2} (no MC effect) to $\epsilon=8.5$ Å^{-2} [above the value $(A-B)^{-2}\sim 6.5$ Å^{-2}]. The quantum-mechanical calculations were done using the method developed especially for the particle-ellipsoid system.[3]

The solid line is the right differential cross section ($\theta>0$) and the crosses are the left ($\theta<0$). We notice that for ϵ below the critical value for the onset of the MC effect, the left and right cross sections are almost identical. The deviation is small and can be attributed to purely quantum effects. The true ϵ for Li-H_2 is between $\epsilon=5$ Å^{-2} and $\epsilon=6.5$ Å^{-2} in the same figures, and by a simple interpolation we notice that for the same system the left and right cross sections are not equal. The most obvious asymmetry is for the transition $0\rightarrow 4$, as seen by comparing the position of the first maxima (the rotational rainbow).

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