Semiclassical Perturbation Scattering by a Rigid Dipole

F.T. Smith, D.L. Huestis, and D. Mukherjee

Stanford Research Institute, Menlo Park, California 94025

and

W.H. Miller

Department of Chemistry, University of California, Berkeley, California 94720

(Received 31 July 1975)

A uniform semiclassical S matrix has been developed for collisions of charged particles with rotating rigid dipoles, with use of first-order perturbation theory. The resulting expression is analytical, depending on tabulated functions, and trivial to calculate; it allows evaluations of quantum transitions in classically forbidden regions, and of quantum interference effects.

In the course of studies of the scattering of electrons and ions by simple polar molecular targets, we have discovered a versatile analytical form for the S matrix in the limit of semiclassical perturbation theory when the anisotopic part of the interaction is dominated by a dipole term.

Cross developed a classical perturbation theory of ion-molecule, including ion-dipole, scattering in a formulation emphasizing angular coordinates and spherical trigonometry. We find it advantageous to make maximum use of angular momentum conservation, formulating the problem in the classical version of a (J, M, j, l) angular momentum coupling, where j is the angular momentum of the target molecule and l is the collisional angular momentum, while J and M refer to the total angular momentum and its projection. [Where the distinction is important, j, l, and J will be used for quantum numbers, and the corresponding classical angular momenta are given by expressions like j (∫ 8 +1/2)h.] We have used the semiclassical S-matrix formulation of Miller, combined with first-order perturbation dynamics. By applying a canonical Hamiltonian transformation, we find the phase of the S matrix to have a contribution not identified by Cross.

The uniform semiclassical form for the S matrix provides an appropriate estimate of quantum effects in the nonclassical tunneling region and at the classical turning points, as well as interference effects in the classically allowed region. In addition, by use of the semiclassical S matrix in (J, M, j, l) coupling followed by quantal recoupling using 6-j symbols and construction of the scattering amplitude using the Wigner rotation matrices, we can obtain the quantal diffraction effects in small-angle scattering.

We consider a problem with a zero-order potential interaction in which j and l are separately conserved:

\[ V_0 = V_0(R). \]  

(1)

R is the radial coordinate associated with the collision. In the plane determined by l, the unperturbed collisional motion follows a classical trajectory given by R(t) and Φ(t), and in the plane determined by J the rotor's unperturbed motion is given by Θ(t). The angle κ between the two planes is fixed by the magnitudes j, l, and J:

\[ N = \cosκ = \frac{\vec{J} \cdot \vec{l}}{||\vec{J}|| ||\vec{l}||} = \frac{(j+\frac{1}{2})^2 + (l+\frac{1}{2})^2 - (J+\frac{1}{2})^2}{2(j+\frac{1}{2})(l+\frac{1}{2})}. \]  

(2)

The line of intersection of the two planes—or the plane perpendicular to it—defines a reference direction for the measurement of Φ and Θ, each in its own plane. It is convenient to take l = 0 at a central point in the trajectory, and assume the unperturbed R(t) to be an even function of t. We can then write Θ(t) = Θ₀ + Θ̄(t) and Φ(t) = Φ₀ + Φ̄(t), where Θ̄(t) and Φ̄(t) are odd functions of t.

The anisotropic interaction will be treated as a perturbation (to which an isotropic part can be added as well). Particular simplifications occur for a dipole interaction, and our attention will be limited to that case; extensions to higher multipoles and nonrigid targets are possible, but they will probably entail greater reliance on numerical methods. We shall assume the general form of the interaction to be

\[ V_1(R, γ) = U(R) \cosγ, \]  

(3)

where

\[ \cosγ = -\cosΦ \cosΘ + N \sinΦ \sinΘ \]

\[ = -\frac{1}{2}(1 + N) \cosψ_+(t) \]

\[ - \frac{1}{2}(1 - N) \cosψ_-(t), \]  

(4)

1073
with
\[
\psi_\pm(t) = \Theta(t) \pm \Phi(t) = \psi_\pm^0 + \overline{\psi}_\pm(t),
\]
and
\[
\psi_\pm^0 = \Theta_0 \pm \Phi_0, \quad \overline{\psi}_\pm(t) = \overline{\Theta}(t) \pm \overline{\Phi}(t).
\]
The dynamic effects of the perturbation can now be evaluated. The associated action integral is a function of \(\Theta_0\) and \(\Phi_0\)
\[
A = A(\Theta_0, \Phi_0) = \int_{-\infty}^{\infty} V_j(R(t), \cos \gamma(t)) \, dt.
\]
This is conveniently simplified through use of the angular variables \(\psi^0_\pm\) and \(\overline{\psi}_\pm(t)\):
\[
A = \overline{A}(\psi^0_+, \psi^0_-) = -\frac{1}{2} (1 + N) B_z \cos \psi^0_+ \]
\[-\frac{1}{2} (1 - N) B_z \cos \psi^0_-,
\]
where
\[
B_z = \int_{-\infty}^{\infty} U(R(t)) \cos \overline{\psi}_+(t) \, dt.
\]
Solving the Hamiltonian equations of motion for the changes in \(l\) and \(j\) to first order, one can easily show that
\[
\hbar \Delta l = -\frac{\partial A(\Theta_0, \Phi_0)}{\partial \Theta_0}, \quad \hbar \Delta j = -\frac{\partial A(\Theta_0, \Phi_0)}{\partial \Phi_0}.
\]
If we define
\[
\Delta k_\pm = \frac{1}{2} (\Delta j \pm \Delta l),
\]
and
\[
Z_\pm = (2\hbar)^{-1} (1 \pm N) B_z,
\]
we have the simple result
\[
\Delta k_\pm = \hbar^{-1} \frac{\partial \overline{A}}{\partial \psi_\pm^0} = -Z_\pm \sin \psi_\pm^0.
\]
The range of classically accessible values of \(\Delta k_+\) and \(\Delta k_-\) is determined by \(Z_\pm\), which depends on the unperturbed values of \(l\) and \(j\), on \(J\), and on the collisional energy \(E\). In addition, each value of \(\Delta k_+\) or \(\Delta k_-\) is associated with a pair of permissible values for the associated angle \(\psi_+^0\) or \(\psi_-^0\), so that each set of values \((\Delta k_+, \Delta k_-)\) can be reached by four different classical trajectories. These will lead to an interference pattern in the \(S\) matrix.

The classical phase is expressed by
\[
\varphi = -\int dt \left[ R'(t) \dot{p}'(t) + \Phi'(t) \frac{\partial V_1}{\partial \Phi} \right],
\]
\[
= \int dt \left[ R(t) \frac{\partial V_1}{\partial R} + \Phi(t) \frac{\partial V_1}{\partial \Phi} + \Theta(t) \frac{\partial V_1}{\partial \Theta} \right],
\]
where \(R', \ p', \ \text{etc.}, \ \text{are "new" variables (related
to the original variables \(R, \ p, \ \text{etc.}, \ \text{through a canonical transformation} \)) chosen in such a way that the coordinates \(R', \ \Theta', \ \Phi'\) appear explicitly only in the perturbation potential term \(V_j\) in the transformed Hamiltonian. With the help of the appropriate canonical transformation, it can be shown that
\[
\varphi = -\Theta_0 \hbar \Delta l - \Phi_0 \hbar \Delta j - A(\Theta_0, \Phi_0) = -\psi_+^0 \hbar \Delta k_+ - \psi_-^0 \hbar \Delta k_- - A(\psi_+^0, \psi_-^0).
\]
Making the appropriate substitutions, we can write this in the form
\[
\varphi = \eta_+ \eta_- + \eta_+ \eta_-,
\]
where
\[
\eta_+ = -\psi_+^0 \Delta k_+ + Z_+ \cos \psi_+^0
\]
\[
= (Z_+^2 - \Delta k_+^2)^{1/2} + \Delta k_+ \sin^{-1} (\Delta k_+ / Z_+). \tag{17}
\]
To construct the \(S\) matrix, we need the Jacobian \(D\) as well as the phase \(\varphi\):
\[
D = \frac{\partial (j, l)}{\partial (\Theta_0, \Phi_0)} = \frac{\partial (\Delta j, \Delta l)}{\partial (\Theta_0, \Phi_0)} = 4 \frac{\partial \Delta k_+}{\partial \psi_+^0} \frac{\partial \Delta k_-}{\partial \psi_-^0} = C_+ C_- \tag{18}
\]
if we take
\[
C_+ = -2 \frac{\partial \Delta k_+}{\partial \psi_+^0} = 2Z_+ \cos \psi_+^0
\]
\[
= 2(Z_+^2 - \Delta k_+^2)^{1/2}. \tag{19}
\]
The symmetry of the solutions is such that nothing is changed if we replace \((\Theta_0, \Phi_0)\) by \((\Theta_0 + \pi, \Phi_0 + \pi)\), which leads to a selection rule of \(\Delta k_+ = \text{integer} \).

The \(S\) matrix is the sum of four terms, corresponding to the four pairs of roots \((\psi_+^0, \psi_-^0)\) allowed by Eq. (13), each being of the form
\[
[(2\pi \hbar)^{1/2} \exp(i\varphi/\hbar)]. \tag{20}
\]
Exercising appropriate care to identify the phases, and making use of the form of \(D\) and \(\varphi\), we find that we can combine terms so that \(S\) can be factored into two independent portions,
\[
S = S_+ S_- \tag{21}
\]
where
\[
S_+ = (4/\pi C_+)^{1/2} \sin(\frac{3}{2} \pi + \beta_+), \tag{22}
\]
with
\[
\beta_+ = \frac{1}{2} C_+ \cos^{-1} (\Delta k_+ / Z_+). \tag{23}
\]
This form is appropriate well into the classical
region of motion, $\Delta k_z^2 < Z_s^2$, or $C_s^2 \gg 0$, Quantum effects become important near $C_s = 0$, and there we can make use of a uniform approximation in the Bessel function form, which reduces to (22) in the limit of large $C_s$:

$$S_z = J_{\Delta k_z}(Z_s).$$  \hspace{1cm} (24)$$

When $C_s$ becomes imaginary, (24) goes over into an exponential tunneling form valid for $\Delta k_z^2 \gg Z_s^2$, or $C_s^2 \ll 0$:

$$S_z = (|C_s|)^{1/2} \exp(-|\beta_s|).$$  \hspace{1cm} (25)$$

The form (24), however, is valid everywhere, and is essential near the edge of the classical region, where $C_s^2 \to 0$, i.e., at the classical turning points.

The $S$ matrix is desired in terms of the initial and final angular momenta, which are connected with $l$, $J$, $\Delta J$, and $\Delta l$ by

$$l_1 = l - \Delta l/2, \quad l_2 = l + \Delta l/2,$$
$$j_1 = j - \Delta J/2, \quad j_2 = j + \Delta J/2.$$  \hspace{1cm} (26)$$

Also the initial and final collisional energies are connected with the average value $E$ by

$$E + (j + \frac{1}{2})\hbar^2/2I = E_1 + (j_1 + \frac{1}{2})\hbar^2/2I = E_2 + (j_2 + \frac{1}{2})\hbar^2/2I.$$  \hspace{1cm} (27)$$

With use of these relations [(26) and (27)] and the properties of the Bessel functions, it is easy to show that this $S$ matrix is correctly symmetric and approximately unitary—approaching unitarity correctly as $l$ and $j$ grow large. It is not difficult to renormalize it for small $j$ and $l$ to make it correctly unitary.

We have evaluated the integrals $B_i$ [Eq. (9)] for two specific cases, in both of which the perturbation is an ion- or electron-dipole interaction,

$$V_i = (\mu e/R^2) \cos \gamma.$$  \hspace{1cm} (28)$$

When the target is a neutral dipole, case (a), the unperturbed potential vanishes, $V_0 = 0$. The result can be expressed in terms of a parameter measuring the ratio of angular velocities of the rotor and the collisional motion at $t = 0$,

$$\omega_a = (j + \frac{1}{2})(l + \frac{1}{2})\hbar^2/2IE,$$  \hspace{1cm} (29)$$

where $I$ is the moment of inertia of the dipole, a factor

$$m\mu e/\hbar^2(l + \frac{1}{2}),$$  \hspace{1cm} (30)$$

and the integrals

$$I_{1,a} = \omega_a [K_1(\omega_a) + K_0(\omega_a)].$$  \hspace{1cm} (31)$$

The same integrals appear in the problem as formulated by Cross and also in Percival’s theory of excitation of hydrogenic atoms by charged particles. Then the quantity $Z_s$ becomes

$$Z_s = (m\mu e/\hbar^2)(l + \frac{1}{2})^{1/2}(1 \pm N)I_{1,a}(\omega_a).$$  \hspace{1cm} (32)$$

We are using this solution in a study of scattering and rotational excitation in collisions of electrons or ions with polar molecules.

When the dipolar target has a net charge, and the bombarding species is an ion, $V_0$ is the Coulomb potential, $V_0 = \pm e^2/R$. We have found an interesting solution for a special case, where the unperturbed collisional energy $E$ is 0 and the trajectory is parabolic. In that case, (b), we have

$$\omega_b = (j + \frac{1}{2})(l + \frac{1}{2})\hbar^2/me^2I,$$  \hspace{1cm} (33)$$

and the integrals $I_{1,b}$ lead to Airy functions,

$$I_{1,b} = 2\pi [x Ai(x) - x^{1/2} Ai'(x)],$$
$$x = (\frac{1}{2}\omega_b^2)^{3/2}.$$  \hspace{1cm} (34)$$

This solution is of interest in connection with capture or detachment between free (hyperbolic) trajectories with eccentricity slightly greater than 1, and large elliptic orbits with eccentricity slightly less than 1.

The separation of the equations connecting the phase angles $\psi_\pm$ with the momenta $\Delta k_z$ in the simple form of Eq. (13) appears to be peculiar to the dipole angular dependence of $V_1$. It is independent of the $R$ dependence of $V_0$ and $V_1$, and so the factorization of the $S$ matrix and the general form (24) of its factors persists, only the integrals $B_i$ or $I_i$ changing with the functional form of the $R$ dependence. These integrals can be obtained by numerical quadrature when an analytical form is not available.

When the angular dependence of $V_1$ is not that of a simple dipole, $P_i(\cos \gamma)$, the elimination of the phase angles and the expression of $S$ as a function of the angular momenta and their perturbations is not so simple as in the dipole case, and may have to be carried out by numerical interpolation. In that case, the $S$ matrix will probably not factorize.

*Work supported by the U. S. Air Force Cambridge Research Laboratories, the National Science Foundation, the U. S. Air Force office of Scientific Research.
Limitation of Brillouin Scattering in Plasmas\textsuperscript{*}

W. L. Kruer, E. J. Valeo, and K. G. Estabrook

*Lawrence Livermore Laboratory, University of California, Livermore, California 94550

(Received 2 June 1978)

It is shown that Brillouin scattering of intense laser light in plasmas can be limited by momentum and energy deposition as a result of reflection of the light. The calculations compare favorably with some recent experimental results.

The efficiency with which intense laser light is absorbed is one of the most important questions for laser-fusion applications. Of particular concern is the possibility that intense light may be reflected in the underdense plasma by the Brillouin and Raman instabilities. Linear theory\textsuperscript{1} indicates that the thresholds for these instabilities are readily exceeded, and computer simulations\textsuperscript{2-5} have shown that a large reflection can then occur in the nonlinear state. Reflection via the Brillouin instability is especially dangerous, since the principal energy transfer is from the incident light wave to the scattered one. However, there is at present very little correlation between experiment and theory. For example, experiments\textsuperscript{6-10} have typically shown a net back reflection of order 20\% for incident light intensities of \( \approx 10^{15} \) W/cm\(^2\) (for 1.06-\mu m light). Indeed this back reflection is sometimes observed to decrease with increasing intensity.

We present theoretical estimates and computer simulations which show that a large induced reflection of intense light can occur, but that this reflection is inherently limited by momentum and energy deposition as a result of reflection of the light in the underdense plasma. This momentum deposition drives a reflection front supersonically through the underdense plasma. Consequently, the induced reflection persists for a limited time which depends on both the intensity and the amount of plasma present in the underdense region. We compare our results with some recent experimental results\textsuperscript{6} obtained at the University of Rochester in which a large and time-dependent reflection has been observed.

The basic physical processes can be simply illustrated by appealing to an idealized model. Assume that the reflection length \( l \) is much less than the density scale length \( L \) of the underdense plasma. Analytic calculations of this reflection length in both the weakly\textsuperscript{11} and the strongly\textsuperscript{12} damped acoustic-wave limits show that this condition is easily obtained. Further assume that the ion-wave-damping decrement satisfies the inequality \( \nu_i > c_s / L \), where \( c_s \) is the sound speed. Then the ion waves deposit their momentum and energy in the interaction region. Computer simulations\textsuperscript{5} have shown that, even if the ion waves are initially weakly damped, they become heavily damped in the nonlinear state because of ion trapping.

Finally, assume that the energy density of the electromagnetic wave exceeds the kinetic-energy density of the plasma. This assumption is motivated by simulation studies\textsuperscript{13} of laser-light absorption. These studies indicate that near the critical density \( n_c \) the plasma electron energy distribution consists of a relatively cold main body with a temperature of \( \sim 1 \) keV due to inverse bremsstrahlung, plus a lower-density very hot tail due to various collective heating mechanisms. Since this lower-density hot tail is rather ineffective in reflecting the light, we neglect it here. For a main body temperature of 1 keV, our last assumption is satisfied when the intensity \( I > 2 \times 10^{15} \) W/cm\(^2\) for 1.06-\mu m light.

With these assumptions, consider the macroscopic behavior in the frame moving with the reflection front. As illustrated in Fig. 1, take total light and plasma reflection from opposite sides of the front, and designate the incoming (reflected) plasma density and velocity by \( n^+ (n^+ \text{ and } v^+) \)