Atomic versus molecular diffraction: Influence of break-ups and finite size

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Abstract

Atomic diffraction through double slits and transmission gratings is well described in terms of the associated de Broglie waves and classical wave optics. However, for weakly bound and relatively large systems, such as the He₂ dimer, this might no longer hold true due to the possibility of break-up processes and finite-size effects. We therefore study the diffraction of weakly bound two-particle systems. If the bar and slit widths of the grating are much larger than the diameter of the two-particle system we recover the usual optics results. For smaller widths, however, deviations therefrom occur. We find that the location of possible diffraction peaks is indeed still governed by the usual grating function from optics, but the peaks may have a lower intensity. This is not unexpected when break-up processes are allowed. More unusually though, diffraction peaks which would be absent for de Broglie waves may reappear. The results are illustrated for diffraction of He₂.

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I. INTRODUCTION

The rapidly developing new field of atom optics [1] exploits wave aspects of quantum mechanical particles and associated interference effects, similar to electron and neutron interferometry. Typical optics experiments have been carried over to atoms. For example, an atomic beam can be passed through a double slit or through a transmission grating and the diffraction pattern can be observed [2]. In such experiments the incoming atoms usually can be considered as point particles and described by plane waves. The standard wave-theoretical methods and approximations of classical optics (Huygens, Kirchhoff) have been successfully applied in atom optics and yield in general good agreement with experiment. Indeed, in view of the success of these methods one might be inclined to think that all theory needed for atom optics is contained in a good optics book.

New problems and challenges, however, might arise for diffraction experiments with molecules, in particular with weakly bound systems. A system, when observed at a nonzero diffraction angle from the incident direction, clearly has received a momentum transfer from the grating. For weakly bound systems this might induce break-up processes which in turn might change the diffraction pattern. Moreover, weakly bound systems in general are larger than atoms. Treatment as a point particle might therefore not be adequate when the grating slits approach the size of the system and when therefore more break-up processes are to be expected. In fact, it is an interesting and difficult dynamical problem how a relatively large molecule squeezes through a slit.

In view of recent diffraction experiments with molecules this is not a moot question. Grating diffraction of both He$_2$ [3] and of Na$_2$ [4] have been observed [5]. The molecule Na$_2$ is very small compared to present-day slit widths and the binding energy is large compared to the incident molecular kinetic energy. Therefore no appreciable deviations from point particle diffraction are expected even if present-day slit widths are reduced by a factor of 2 or 4. On the other hand, for the helium dimer He$_2$, discovered a few years ago [6,3], the extremely low binding energy of $-0.11$ meV (see e.g. Ref. [7]) is much smaller than the incident kinetic energy of typical experiments. Its estimated diameter of about 6 nm [8] is still an order of magnitude smaller than the slit widths used in the experiments, but a further reduction in the slit width might make a difference.

A theoretical analysis of diffraction of weakly bound systems by a transmission grating, as undertaken in this paper, may therefore be of more than only academic interest. We restrict ourselves to diatomic systems. Since diffraction by a transmission grating is really a scattering by the bars of the grating and since we want to include break-up processes and finite-size effects we use quantum mechanical scattering theory. As in the case of point-particle diffraction we describe the effect of the grating bars by a short-range repulsive (“reflecting”) potential. Attractive parts to the potential could also be included, but for simplicity this is not done here.

As usual we neglect the electronic degrees of freedom of the two atoms. We thus consider a bound system of two interacting particles and its scattering by an external potential. Due to the presence of the latter this problem does not resemble so much one-particle scattering but rather the three-body problem (the external potential playing the role of the third body). Not surprisingly one can therefore use similar methods. We have found it advantageous to employ the Faddeev approach [8] in its formulation by Alt-Grassberger-Sandhas (AGS) [10].
Briefly, our results are the following. We obtain an analytic expression for the elastic diffraction amplitude of a weakly bound two-particle system which – implicitly – incorporates break-up processes and finite-size effects. We also give an expression for diffraction including excitation and de-excitation of the two-particle system, but the break-up channel is not explicitly treated here.

Provided the bar and slit width of the grating are not much smaller than the molecular diameter, the diffraction amplitude contains the familiar grating function of classical optics as a factor. This grating function appears automatically from our general results, and we give a simple intuitive argument why its appearance is not surprising. The grating function, which just as in classical optics determines the peaks at the different diffraction orders, is multiplied by the molecular diffraction amplitude due to a single bar of the grating, and this is where a difference to the point-particle case may arise.

We show that for a molecular diameter very small compared to the bar and slit width the deviations from the point-particle result are indeed negligible, as expected. This then verifies the interpretation of the He\textsubscript{2} diffraction curves given in Refs. [3,11]. For smaller bar and slit widths noticeable deviations occur. In particular, heights of diffraction peaks may become lower than for a point particle. This can clearly be attributed to break-ups which diminish the actual number of molecules in a given direction. Moreover, the decrease in peak height goes with the diffraction angle and thus with the lateral momentum transfer. This is also intuitively clear since higher momentum transfer implies more break-ups. As another effect, diffraction orders which are sometimes suppressed to zero for point particles may reappear. This is probably a finite-size effect.

If the slits become much smaller than the diameter of the two-particle system, additional terms appear in the diffraction pattern. This case, however, is presently not of experimental interest and not further evaluated here.

The plan of the paper is as follows. In Section II we establish AGS equations for the scattering amplitude of a bound two-particle system for scattering by an external potential. These equations decouple in our case. They can be iterated in a straightforward way, and in Section III we specialize the lowest order term to diffraction scattering. In Section IV we evaluate this for the diffraction of a weakly bound two-particle system by a transmission grating. The reader not interested in the detailed derivation can proceed directly to the main result in Eqs. (46) to (48). As an illustration of our general formulae, in Section V diffraction patterns for He\textsubscript{2} are determined for three slit widths and compared with the corresponding patterns for a point particle. A full discussion of the He\textsubscript{2} case and a detailed comparison with corresponding experimental results will appear elsewhere [12].

II. GENERAL THEORY

We consider a bound two-particle system – designated as (diatomic) “molecule” – which is scattered by an obstacle. Later we will specialize to diffraction by a transmission grating. The two particles are taken to represent two atoms whose electronic degrees of freedom are neglected. The (molecular) binding potential of the two particles is denoted by $V$ which may have several bound states, with (negative) binding energy $E_\gamma$ and wave function $\phi_\gamma(x)$ where $x \equiv x_1 - x_2$ and $x_1$ and $x_2$ are the coordinates of the two particles. The obstacle (grating) is represented by a potential $W$ of the form
\[ W(\mathbf{x}_1, \mathbf{x}_2) = W_1(\mathbf{x}_1) + W_2(\mathbf{x}_2) \]  

where \( W_1 \) and \( W_2 \) are short-range and strongly repulsive \(^{[3]}\).

For a molecule, as opposed to a point particle, there are various scattering channels. In the elastic channel the molecule is scattered without energy transfer, while in the inelastic channels the molecule might end up in a different bound state or might break up. In a break-up process both constituents may emerge as asymptotically free particles or one of them may stick to the obstacle if \( W \) has an attractive part.

The total Hamiltonian for the problem is \( H_0 + V + W \), with \( H_0 \) the kinetic energy,

\[ H_0 \equiv \frac{\hat{p}_1^2}{2m_1} + \frac{\hat{p}_2^2}{2m_2} = \frac{\hat{P}^2}{2M} + \frac{\hat{p}^2}{2\mu} \]  

where \( \hat{p}_1 \) and \( \hat{p}_2 \) are the momentum operators of the particles, \( \hat{P} \) and \( \hat{p} \) those for the total and relative momentum, and \( M \) and \( \mu \) the total and reduced mass, respectively. For elastic and (de-)excitation scattering the channel Hamiltonian is \( H_0 + V \). Its improper eigenstates are denoted by \( |\mathbf{P}, \phi_\gamma\rangle\),

\[ (H_0 + V)|\mathbf{P}, \phi_\gamma\rangle = (\frac{\hat{P}^2}{2M} + E_\gamma)|\mathbf{P}, \phi_\gamma\rangle \equiv E|\mathbf{P}, \phi_\gamma\rangle. \]  

As in the case of one-particle scattering the \( S \) matrix can be decomposed as

\[ \langle \mathbf{P}, \phi_\gamma|S|\mathbf{P}', \phi_\gamma'\rangle = \delta_{\gamma\gamma'}\delta^{(3)}(\mathbf{P} - \mathbf{P}') - 2\pi i\delta(E - E')t(\mathbf{P}, \phi_\gamma; \mathbf{P}', \phi_\gamma') \]  

where \( t(\mathbf{P}, \phi_\gamma; \mathbf{P}', \phi_\gamma') \) is the transition amplitude for an incoming molecule of momentum \( \mathbf{P}' \) and internal state \( \gamma' \) to an outgoing one of momentum \( \mathbf{P} \) and internal state \( \gamma \).

In one-particle scattering the scattering amplitude can usually be determined by means of a Lippmann-Schwinger (LS) equation. In the present multi-channel situation there is a set of coupled LS equations, and one meets the same difficulties with them as in the three-body problem. In the latter one therefore uses the Faddeev approach \(^{[3]}\). For the calculation of transition amplitudes one often employs its formulation by Alt-Grassberger-Sandhas (AGS) \(^{[10]}\). It turns out that the AGS approach is easily carried over to the present two-body problem with its external potential. Moreover, the resulting equations decouple and can be evaluated for interesting situations. This will now be outlined.

We introduce the Green’s operators

\[ G_0(z) \equiv (z - H_0)^{-1} \]  
\[ G(z) \equiv (z - H_0 - V - W)^{-1} \]  
\[ G_V(z) \equiv (z - H_0 - V)^{-1} \]  

and similarly \( G_W(z) \). Here \( G_0 \) is the free and \( G \) the total resolvent. One has the usual resolvent equations

\[ G = G_V + G_V W G \]  
\[ G = G_W + G_W V G. \]
\[ T_V(z) \equiv V + V G_V(z) V \]
\[ T_W(z) \equiv W + W G_W(z) W \]

which are the \( T \) matrices \[14\] for the potential \( V \) and \( W \), respectively, in the two-particle Hilbert space. They satisfy

\[ T_V G_0 = V G_V \]
\[ T_W G_0 = W G_W \]

which follows by writing Eq. \[8\] as

\[ T_V = V G_V (G_V^{-1} + V) = V G_V G_0^{-1} \]

and similarly for \( T_W \).

In analogy to the treatment of the three-body case in Ref. \[15\] we now introduce operators \( U_{VV}(z) \) and \( U_{WW}(z) \) through the equations

\[ G(z) = G_V(z) + G_V(z) U_{VV}(z) G_V(z) \]
\[ G(z) = G_W(z) U_{WW}(z) G_V(z). \]

Proceeding verbatim as in Section 7 of Ref. \[15\] one shows that the transition amplitudes of Eq. \[4\] are given by

\[ t(P, \phi; P', \phi') = \langle P, \phi | U_{VV}(E + i0) | P', \phi' \rangle \]

where

\[ E = P'^2/2M + E_{\gamma'} = P^2/2M + E_{\gamma}. \]

Thus \( U_{VV} \) acts as a transition operator, and its advantage is that it satisfies a simple equation, seen as follows. We equate Eqs. \[8\] and \[13\],

\[ G_V + G_V W G = G_V + G_V U_{VV} G_V, \]

and insert Eq. \[14\] for \( G \). This gives

\[ U_{VV} = W G_W U_{WW} = T_W G_0 U_{WW} \]

by Eq. \[11\]. Similarly equating Eqs. \[9\] and \[14\] and inserting Eq. \[13\] gives

\[ U_{WW} = G_0^{-1} + T_V G_0 U_{VV}. \]

Eqs. \[18\] and \[19\] are the AGS equations for our problem which decouple here. Indeed, insertion of Eq. \[19\] into Eq. \[18\] gives

\[ U_{VV} = T_W + T_W G_0 T_V G_0 U_{VV} \]

which no longer contains \( U_{WW} \). This is an exact equation for the transition operator and for the transition amplitudes in Eq. \[15\]. Break-up processes and finite-size effects are exactly included.
Similar to Eq. (15) one can show that

\[ U_{0V} \equiv G_0^{-1} + T_V G_0 U_{VV} + T_W G_0 U_{WV} \]  

(21)

is the transition operator for break-up processes into two asymptotically free particles. Thus a knowledge of \( U_{VV} \) and \( U_{WV} \) would also cover these processes. One can further show that Eqs. (18) and (19) yield unitarity of the \( S \) matrix.

The great advantage of Eq. (20) or of the system in (18) and (19) is that it lends itself immediately to a perturbative approach by iteration. Such an iterative procedure is also used in the multiple-scattering expansion in the three-body problem [14]. To lowest order one has

\[ U_{VV} \approx T_W \]  

(22)

and as in Ref. [14] this is expected to give a good approximation for large incident energy, viz. for

\[ P'^2 / 2M \gg |E_g|, \langle \phi_g | V | \phi_g \rangle \]  

(23)

where \( P' = |P'| \), \( E_g \) is the ground-state energy and \( \phi_g \) the ground-state wave-function. Then in general the above condition also holds for other bound states. The relevant matrix elements of \( T_V \) will then be small compared to the incident energy and this allows one to show that the next order term in the iteration can be neglected. Thus in the region of large incident energy one has, by Eq. (15) for the transition amplitude in Eq. (4),

\[ t(P, \phi_\gamma; P', \phi_\gamma') \approx \langle P, \phi_\gamma | T_W (E + i0) | P', \phi_\gamma' \rangle. \]  

(24)

Although \( T_W \) is the transition operator for scattering of two asymptotically free particles by \( W \), the above matrix element differs crucially from a scattering amplitude for free particles since the wave functions \( \phi_\gamma \) and \( \phi_\gamma' \) do not have a definite (relative) momentum. It is therefore through these wave functions that the above expression takes the interaction potential \( V \) between the two particles and the finite size of the molecule into account.

The major problem remaining is the explicit evaluation of Eq. (24) for the transition amplitude, and this we will do here for the experimentally interesting case of diffraction by a grating and for small scattering angles.

## III. SPECIALIZATION TO DIFFRACTION SCATTERING

The operator \( T_W(z) \) appearing in the approximate expression for the transition amplitude in Eq. (24) is, in principle, the \( T \) matrix for the scattering of two asymptotically free particles by the potential \( W = W_1 + W_2 \). But even for this simple additive potential it is not easy to calculate \( T_W(z) \) since (i) it is not evaluated on the energy shell of two asymptotically free particles and (ii) the Green’s operator \( G_W \) is not easily expressed by one-particle Green’s operators. We therefore restrict ourselves in the following to the experimentally important diffraction domain and to small scattering angles.

In the diffraction domain the de Broglie wave-length is, by definition, small compared to the extension of the obstacle and the potential [16]. Small scattering angles mean \( |P_\perp| \ll \)
\( P' \) where \( P_\perp \) is the component of \( P \) orthogonal to the incident momentum \( P' \). For a transmission grating of \( N \) bars, grating period \( d \) and slit width \( s \) the domain of diffraction scattering can be characterized by the condition
\[
P's/\hbar, P'(d - s)/\hbar \gg 1.
\]
We also assume here and in the following that the masses \( m_1 \) and \( m_2 \) are of the same order of magnitude.

To calculate the matrix element of \( T_W \) in Eq. (24) one could proceed as in the eikonal approximation [16] by considering an absorbing (“black”) obstacle, instead of a reflecting one, and thus work with a complex ersatz potential instead of the real potential \( W \). We have done this, and the results agree with those of the more direct approach we are presenting here.

Inserting two complete sets of two-particle plane waves in the r.h.s. of Eq. (24) and performing the integration over resulting \( \delta \) functions one obtains
\[
\langle P, \phi_\gamma | T_W (E + i0) | P', \phi_\gamma' \rangle = \int d^3p d^3p' \phi_\gamma^* (p) \times \langle p_1, p_2 | T_W (E + i0) | p'_1, p'_2 \rangle \phi_\gamma' (p')
\]
where for \( p_1 \) and \( p_2 \) one has to insert
\[
p_{1,2} = \frac{m_{1,2}}{M} P \pm p
\]
and similarly for \( p'_{1,2} \). We note that the kinetic energies of \( |p_1, p_2| \) and \( |p'_1, p'_2| \) are not equal to \( E \) so that we are not on the energy shell. However, in Eq. (27) only those values of \( p \) and \( p' \) enter for which \( \phi_\gamma (p) \) and \( \phi_\gamma' (p') \) are essentially different from zero. The condition Eq. (23) of large incident energy ensures that \( \phi_\gamma (p) \) and \( \phi_\gamma' (p') \) are centered at momenta which are small compared to \( P' \) since the Schrödinger equation implies, upon multiplication with \( \langle \phi_\gamma, \langle \hat{p}^2 | \phi_\gamma \rangle /2\mu = E_\gamma - \langle \phi_\gamma | V | \phi_\gamma \rangle \ll P'^2 /2M. \)

Therefore one has indeed
\[
|p|, |p'| \ll P' \approx |P| \equiv P
\]
and thus, by Eq. (27),
\[
p'_{1,2} \equiv |p'_{1,2}| \approx \frac{m_{1,2}}{M} P' \quad p_{1,2} \equiv |p_{1,2}| \approx \frac{m_{1,2}}{M} P.
\]
Hence
\[
E'_1 + E'_2 \equiv \frac{p'^2}{2m_1} + \frac{p'^2}{2m_2} \approx P'^2 /2M \approx E
\]
where in the last step Eqs. (13) and (23) have been used. Now one can convince oneself by explicit calculation that under the above conditions \( \langle p_1, p_2 | T_W (E + i0) | p'_1, p'_2 \rangle \) is slowly varying in \( E \) so that \( E \) can be replaced by the l.h.s. of Eq. (31). Hence
\[ \langle p_1, p_2 | T_W (E + i0) | p_1', p_2' \rangle \approx \langle p_1, p_2 | T_W (p_1'^2 / 2m_1 + p_2'^2 / 2m_2 + i0) | p_1', p_2' \rangle. \] (32)

We now use the LS equation for the potential \( W \) and the corresponding two-particle scattering states \( |p_1', p_2', +\rangle \),

\[ |p_1', p_2', +\rangle = |p_1', p_2'\rangle + G_W (p_1'^2 / 2m_1 + p_2'^2 / 2m_2 + i0)W |p_1', p_2'\rangle. \] (33)

Using Eq. (10) for \( T_W \), pulling out a factor of \( W \) and then using the LS equation we obtain

\[ \langle p_1, p_2 | W | p_1, p_2 \rangle \] (27),

and correspondingly for \( W \)

the one-particle amplitudes

\[ \langle p_1 | W | p_1 \rangle \]

and similarly for particle 2. In this way the r.h.s. of Eq. (34) can be written in terms of

\[ \langle p_1 | p_1' + \rangle_1 \]

for \( W_1 \) and \( W_2 \), respectively, and \( |p_1, p_2\rangle = |p_1\rangle |p_2\rangle \). We thus have from Eq. (32)

\[ \langle p_1, p_2 | T_W (E + i0) | p_1', p_2' \rangle \] (34)

To evaluate this we introduce one-particle amplitudes

\[ t_1(p_1, p_1') \equiv \langle p_1 | W_1 | p_1' \rangle_1 \] (35)

and similarly for \( t_2(p_2, p_2') \). For the special case \( p_i^2 = p_i'^2 \) these amplitudes would just be physical transition amplitudes \([17]\), but in general they are off the energy shell. We also need \( \langle p_1 | p_1', + \rangle_1 \), and for this we use the LS equation

\[ |p_1', +\rangle_1 = |p_1'\rangle + (E_1' - p_1'^2 / 2m_1 + i0)^{-1}W_1 |p_1', +\rangle_1 \] (36)

where \( E_1' = p_1'^2 / 2m_1 \) in the free one-particle Green’s operator. One obtains

\[ \langle p_1 | p_1', + \rangle_1 = \delta(3)(p_1 - p_1') + \frac{t_1(p_1, p_1')}{E_1' - E_1 + i0} \] (37)

and similarly for particle 2. In this way the r.h.s. of Eq. (34) can be written in terms of

the one-particle amplitudes \( t_1 \) and \( t_2 \) and inserted into Eq. (24). We have thus completely expressed the transition amplitude in Eq. (24) in terms of one-particle amplitudes.

To further evaluate the integrals over \( d^3p \) and \( d^3p' \) in Eq. (26) we use the condition of large incident momentum and small scattering angles. As outlined above this makes \( p \) small in Eq. (27), and correspondingly for \( p' \). A simple calculation then shows that

\[ \frac{1}{E_1' - E_1 + i0} + \frac{1}{E_2' - E_2 + i0} \approx -\frac{2\pi iM}{P} \delta(p_\parallel - p_\parallel') \] (38)

where \( \parallel \) denotes the component in the incident direction, i.e. parallel to \( P' \). In order to make Eq. (38) applicable we use that \( t_i(p_i, p_i') \) depends only weakly on \( p_\parallel \) and \( p_\parallel' \) since in Eq. (27) for \( p_i \) and \( p_i' \) one has \( |p'|, |p| \ll P', P \), by Eq. (29) \([18]\). With Eq. (38) an elementary calculation then gives for the transition amplitude

\[ t(P, \phi; P', \phi') \] (39)

\[ \times t_2(P - P' + \frac{m}{M}P' - p', \frac{m}{M}P' - p') \phi_{\parallel}(p') \]

\[ + \int d^3p' \phi_{\parallel}^*(p' + \frac{m}{M}P - p') \phi_{\parallel}(p') \]

\[ - \frac{2\pi iM}{P} \int d^2p_\perp \int d^3p' \phi_{\parallel}^*(p'|p_\perp) t_1(\frac{m}{M}P + p_\parallel, \frac{m}{M}P + p_\parallel, \frac{m}{M}P' + p') \]

\[ \times t_2(\frac{m}{M}P_\perp - p_\parallel', \frac{m}{M}P_\perp - p_\parallel', \frac{m}{M}P' - p') \phi_{\parallel}(p'). \]
This result holds in the diffraction domain, for small scattering angles and for \( m_1 \) and \( m_2 \) of the same order of magnitude.

**IV. EVALUATION FOR DIFFRACTION GRATINGS**

We now consider small-angle diffraction of a weakly bound two-particle system (molecule) by a transmission grating in normal incidence (see Fig. 1). By \( \perp \) we denote the component of a vector orthogonal to \( \mathbf{P}' \) and by \( \parallel \) the component parallel to \( \mathbf{P}' \). Since the diffraction condition and \( |p'|, |p| \ll P', P \) hold, by Eq. (29), the one-particle amplitudes appearing in Eq. (39) are only weakly dependent on \( p_\parallel \) and \( p'_\parallel \). One can therefore take them on-shell. Under our conditions on \( p_i \) and \( p'_i \) the one-particle amplitudes are then of the form

\[
t_i(p_i; p'_i) \equiv t(p_i/m_i; p_i - p'_i). \tag{40}
\]

An explicit expression will be given later, but the dependence on the lateral momentum transfer already allows a simplification of Eq. (39). Using \( p_i/m_i \approx P/M \), by Eqs. (29) and (27), one obtains in a straightforward way

\[
t(P, \phi_{\gamma'}; P', \phi_{\gamma'}) = t(P/M; m_1 M p_\perp) \left\{ F_{\gamma'\gamma}(q) + F_{\gamma'\gamma}^*(q) \right\} \tag{41}
\]

where

\[
F_{\gamma'\gamma}(q) \equiv \int d^3x \phi_{\gamma}(x) e^{iq \cdot x/h} \phi_{\gamma'}(x). \tag{42}
\]

For \( \gamma = \gamma' \) the latter is the Fourier transform of \(|\phi_{\gamma}(x)|^2\). \( F_{\gamma\gamma}(q) \) is even and real, and it could be called a molecular form factor. Again we recall that we have assumed diffraction scattering, small angles, and \( m_1 \) and \( m_2 \) of the same order of magnitude [19].

From now on we consider elastic scattering (\( \gamma = \gamma' \)). For a transmission grating of \( N \) bars as in Fig. 1 the one-particle amplitude in Eq. (40) is calculated for reflecting bars by standard methods [20,21] as

\[
t(p_i/m_i; q_\perp) = H(q_2) t^\text{PP}_{\text{bar}}(p_i/m_i; q_2) \delta(q_3) \tag{43}
\]

where \( q_\perp \equiv p_i - p_i' \), \( H \) is the usual grating function [22]

\[
H(q_2) = \frac{\sin(q_2 N d/2h)}{\sin(q_2 d/2h)} \tag{44}
\]

and where \( t^\text{PP}_{\text{bar}} \) is the point-particle amplitude [23] for a single bar (in dimension two),

\[
t^\text{PP}_{\text{bar}}(p_i/m_i; q_2) = -\frac{2i p_i \sin[q_2(d - s)/2h]}{2\pi} \frac{\sin[q_2(d - s)/2h]}{q_2}. \tag{45}
\]

The \( \delta \) function appears here because we have taken the bars from \(-\infty \) to \( \infty \) in the third direction. As a consequence of this the molecular transition amplitude in Eq. (41) is a
product of $\delta(P_3)$ and of a smooth function, and the norm-square of the latter is proportional to the experimentally relevant differential cross section.

To further evaluate the transition amplitude we first consider the simplest case where both the bar and slit width are much larger than the diameter of the two-particle system. Then the one-particle amplitude $t(P/M;P_{\perp})$ entering Eq. (41) is sharply peaked around $P_{\perp} = 0$, on a scale for which $F_{\gamma\gamma}(m_1P_{\perp})$ will in general vary only very little and hence one can replace it by $F_{\gamma\gamma}(0) = 1$. A simple calculation then shows that the integral in Eq. (41) cancels half of the first term. The net result is $t(P/M;P_{\perp})$. This is just the result for a point particle of mass $M = m_1 + m_2$ and incident and final momentum $P'$ and $P$, respectively, yielding the diffraction pattern of classical optics for wave numbers $k' = P'/\hbar$ and $k = P/\hbar$. Thus in the case of bar and slit width much larger than the diameter of the two-particle system one recovers for the elastic case the expected point-particle result [24].

The situation changes when bar and slit width decrease. The grating function $H$ in Eq. (44) can be written in an elementary way as a geometric sum of $\exp\{-iq_2d/\hbar\}$, up to a phase factor. Inserting this into Eq. (43) and subsequently into Eq. (41) then yields for the molecular transition amplitude a simple sum plus a double sum from the integral term. Combining the diagonal part of the double sum with the first sum gives what we call the coherent part $t_{\text{coh}}$ of the amplitude, while the nondiagonal remainder of the double sum gives the incoherent part,

$$t(P,\phi_\gamma;P',\phi_\gamma) = t_{\text{coh}}(P_{\perp}) + t_{\text{incoh}}(P_{\perp}).$$

(46)

The coherent part can be interpreted as the contribution from those processes where the diatomic molecule interacts with a single bar, while the incoherent part contains the interaction with different bars. Provided $d$ and $s$ are not too small, i.e. still comparable to the molecular diameter, then $t_{\text{incoh}}$ can be shown to be negligible and an elementary calculation gives for the coherent part

$$t_{\text{coh}}(P_{\perp}) = t_{\text{mol}}(\gamma, P/M;P_2)H(P_2)\delta(P_3)$$

(47)

where $H$ is the grating function of Eq. (14) and $t_{\text{mol}}$, the transition amplitude for a single bar of width $d - s$, is given by

$$t_{\text{mol}}(\gamma, P/M;P_2) = t_{PP}(P/M;P_2)\int d^3x \left\{ e^{iP_2x_2/M} + e^{iP_2x_2/M} \right\} |\phi_\gamma(x)|^2$$

$$+ \frac{2iP}{(2\pi)^2M} \int dx_1dx_3\int_0^{d-s}dx_2 |\phi_\gamma(x)|^2$$

$$\times \left\{ \sin \left[ \frac{P_2x_2}{\hbar} \left( \frac{d-s}{2} - \frac{m_2x_2}{M} \right) \right] + \sin \left[ \frac{P_2x_2}{\hbar} \left( \frac{d+s}{2} - \frac{m_1x_2}{M} \right) \right] \right\}/P_2,$$

(48)

with $t_{PP}$ as in Eq. (13) [21]. These results for elastic diffraction by gratings hold for small diffraction angles, for strongly repulsive short-range bar potentials (“reflecting” bars) and for the two constituent masses of comparable magnitude. If bar and slit width become much smaller than the diameter of the two-particle system, then $t_{\text{incoh}}$ can no longer be neglected.

The grating function $H$ in Eq. (17) with its sharp peaks is the same as for diffraction of point particles [25]. However, instead of $t_{PP}$ one has now $t_{\text{mol}}$ in which the bound-state wave-function of the diatomic molecule enters, and this may change the height of the peaks. Indeed, physically one would expect break-up processes to diminish the number of bound systems scattered into a given direction. This does indeed happen for certain diffraction
peaks, as will be seen in the example of the next section. However, also the reverse can happen, as seen by the following simple argument for equal bar and slit width \((s = d/2)\). In this case the point particle amplitude \(t_{PP}^{\text{bar}}\) for a single bar has a zero at the even-order peaks of the grating function and hence these diffraction peaks vanish for point particles. But in the molecular case \(t_{\text{mol}}^{\text{bar}}\) need not be zero there so that even-order diffraction peaks may reappear. This behavior is not so easily understood in terms of break-up processes and is probably a finite-size effect.

V. EXAMPLE: DIFFRACTION OF HELIUM DIMERS

Our result in Eqs. (47) to (48) for the diffraction of weakly bound two-particle systems by a transmission grating can be directly applied to the helium dimer \(\text{He}_2\). First of all, the binding energy of \(\text{He}_2\) is very small, \(E_b/k_B \approx -1.3\) mK (see e.g. Ref. [7]). There seems to be only a single bound state and the estimated bond length of about 6 nm is enormous [8]. The helium dimer \(\text{He}_2\) is by far the largest of known diatomic molecules when considered in their ground state [26]. Our assumption of a totally reflecting one-particle potential seems to be well suited to \(\text{He}_2\) [8]. The bound-state wave-function \(\phi_b(x) \equiv \phi_b(r)\) is rotationally symmetric \((s\) state). For our numerical calculations we employ the analytical expression of \(\phi_b(r)\) given in Ref. [27]. Rotational symmetry allows one to simplify the molecular transition amplitude \(t_{\text{mol}}^{\text{bar}}\) for a single bar in Eq. (48) and to express the multiple integrals as a sum of integrals over \(r\). In Fig. 2 we have evaluated \(|t_{\text{mol}}^{\text{bar}}|^2\) for \(\text{He}_2\) as a function of the lateral momentum transfer \(P_2\) divided by \(\hbar\) (“wave number” \(k_2\)) for various bar widths and have compared it with \(|t_{PP}^{\text{bar}}|^2\) for the corresponding point particle of equal mass as \(\text{He}_2\) (dashed line, from Eq. (45)).

For a grating period of \(d = 100\) nm and slit width \(s = 50\) nm there is little difference between \(t_{\text{mol}}^{\text{bar}}\) and \(t_{PP}^{\text{bar}}\) for lateral momentum transfer up to the fifth order diffraction peak of the grating, and so the overall diffraction pattern in Fig. 3a agrees well with that of a point particle [24]. Since in this case the diameter of \(\text{He}_2\) is small compared to the slit width this verifies the result of the discussion in the preceding section. For \(d = 50\) nm and \(s = 25\) nm the difference in Fig. 2b is more pronounced. First of all, at the odd-order peaks of the grating function – which correspond to the maxima of \(|t_{PP}^{\text{bar}}|^2\), to excellent approximation – \(|t_{\text{mol}}^{\text{bar}}|^2\) is smaller than \(|t_{PP}^{\text{bar}}|^2\) and hence the resulting diffraction peaks should be smaller. This is indeed seen in Fig. 3b and can be attributed to break-ups. Second, \(t_{\text{mol}}^{\text{bar}}\) is nonzero at the zeros of \(t_{PP}^{\text{bar}}\) and hence, by the discussion of the preceding section, the even-orders diffraction peaks should start to reappear. Again this is seen in Fig. 3b. Both phenomena become more and more pronounced for bar and slit width still closer to the \(\text{He}_2\) diameter. This is seen in Figs. 2c and 3c.

A more detailed study of \(\text{He}_2\) diffraction and comparison with experimental results is under way [12].

VI. CONCLUSIONS

In atom optics the finite size of the atoms can usually be ignored, and for instance atom diffraction by a transmission grating is exceedingly well described by matter waves
for a point particle, using the formulae of classical wave optics. The situation may already change for a weakly bound and relatively large molecule of two atoms, such as the helium dimer He$_2$, which is the largest presently known diatomic molecule (in ground state) and which is of great current experimental interest [8,28]. A molecular diameter which is no longer tiny compared to the slit width may have an effect on the diffraction pattern, and the same holds for possible break-ups of a weakly bound system.

In this paper we have studied diffraction scattering of a weakly bound two-particle system, such as He$_2$, and in particular diffraction by a transmission grating for small diffraction angles. The relevant formulae for the transition amplitude for elastic diffraction scattering are given in Eqs. (46) to (48) from which the diffraction pattern is obtained. Insofar as the same grating function appears, which determines the allowed diffraction peaks, it has a structure similar to that for waves which one associates with point particles. This grating function is now multiplied, however, by the transition amplitude for a single bar, and for a diatomic molecule this may differ from that for a point particle. If the bar and slit widths of the grating are much larger than the molecular diameter there is practically no difference to point particles. For smaller bar and slit widths the finite size of the molecule and possible break-up processes may have noticeable effects. For example, as expected in the presence of break-ups, diffraction peaks may have lower height. But, somewhat more unusual, diffraction peaks which are suppressed for point particles may reappear. This is probably due to finite-size effects.

Our derivation used techniques from the three-body problem, in particular the Alt-Grassberger-Sandhas equations, to express the transition amplitude in terms of matrix elements of a transition operator. Their evaluation can be performed with or without the eikonal approximation, and the results for both cases agree.

We intend to carry the quantum mechanical methods employed here over to weakly bound three-particle systems, such as the helium trimer He$_3$. This is an interesting system, in particular in connection with Efimov states [29], and presently under experimental study [28].

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the sin in Eq. (45) and by noting that for \( k_2 = \pm 2\pi n/d, n = 1, 2, \ldots \), i.e. for the main maxima of the grating function, one has \( \sin(k_2d/2) = 0 \) and \( \cos(k_2d/2) = (-1)^n \). We also note that in optics one usually considers gratings which are closed at the two ends and not open as ours. This leads to a change in the zeroth-order peak.

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FIGURES

FIG. 1. Diffraction grating with grating period $d$ and slit width $s$.

FIG. 2. Single-bar transition amplitudes for diatomic molecule and point particle: $|t_{\text{mol}}|^2$ (solid line) and $|t_{\text{bar}}^{\text{PP}}|^2$ (dashed line) for a) $d = 100$ nm, b) $d = 50$ nm, and c) $d = 25$ nm ($s = d/2$). The difference increases for decreasing bar and slit width.

FIG. 3. Calculated helium dimer diffraction pattern (solid line) and respective point-particle pattern (dashed line) for a) $d = 100$ nm, b) $d = 50$ nm, and c) $d = 25$ nm ($N = 30$, $s = d/2$). For smaller bar and slit width the deviation from the point-particle result is increasingly pronounced (see also enlarged insets). In b) and c) the second-order peak noticeably reappears at about $25/(100$ nm) and $50/(100$ nm), respectively.
d = 100 nm (N=30)
d=50 nm (N=30)
d=25 nm (N=30)