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Formation of a molecule by atoms incident upon an external potential

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Two particles incident upon an external potential can combine to form one particle with two components, either by transmission past the potential or by reflection. An example is two atoms combining to form a molecule. The probability of forming a molecule depends sensitively on the strength of the external potential. Delta barrier potentials are shown to be good approximations to narrow Gaussian barriers. Multiple barriers can increase the probability of formation of a molecule up to about 0.5 for reflection and 0.25 for transmission.

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I. INTRODUCTION

We show that two particles incident upon an external potential can combine to form one particle with two components, either by transmission past the potential or by reflection. More specifically, two atoms incident upon an external potential can combine to form a molecule.

Quantum tunneling of particles has been extensively investigated since the beginning of quantum mechanics. The tunneling of diatomic molecules and other composite particles is, however, a fairly new area of research. The earliest work, by Saito and Kayanuma [1], investigated the resonant tunneling of a pair of bound particles incident upon a single barrier. Pen'kov extended this work by considering more general barriers and by proposing a mechanism for the appearance of resonances [2,3]. Resonant tunneling of a pair of bound particles in a one-dimensional lattice was reported by Bulatov and Kornilovitch [4]. Bertulani, Flambaum, and Zelevinsky found the probability of tunneling of a pair of bound particles to be strongly affected by intrinsic structure (e.g., spin) of the pair [5]. Bacca and Feldmeier [6], and also Lee [7], investigated resonant tunneling of a pair of bound particles having binding potentials that are applicable to nuclear physics. Tunneling of a diatomic molecule incident upon a potential barrier in one and three dimensions was reported in Refs. [8,9].

Recent technological advancements have extended the study of quantum tunneling phenomena and have generated experimental applications, including, for example, the tunneling of a single hydrogen atom on a metal surface, which was observed directly by Lauhon and Ho using a scanning tunneling microscope [10]. More exotic examples include resonant tunneling of Cooper pairs [11] and the direct observation of tunneling in a single bosonic Josephson junction [12].

II. FORMULATION OF THE PROBLEM

The Hamiltonian for two atoms, each of mass m, incident upon a potential barrier in one dimension, is

$$H = -\frac{\hbar^2}{2m}\left(\frac{1}{2}\frac{\partial^2}{\partial x^2} + 2\frac{\partial^2}{\partial \xi^2}\right) + V\left(x + \frac{1}{2}\xi\right) + V\left(x - \frac{1}{2}\xi\right) + V_0(\xi), \tag{1}$$

where $\xi \equiv x_1 - x_2$ and $x \equiv \frac{1}{2}(x_1 + x_2)$ are the relative and center of mass (c.m.) coordinates, x_1 and x_2 are the coordinates of the atoms, $V(x_j)$ (for $j=1,2$) is the potential barrier, and $V_0(\xi)$ is the interaction potential between the atoms. We may regard the interacting atoms as being in an unbound molecular state. The wave function $\Psi(x,\xi)$ of a molecule with bound and discrete unbound states, has the form

$$\Psi(x,\xi) = \sum_n \psi_n(x)\chi_n(\xi) + \sum_q \psi_q(x)\chi_q(\xi) = \sum_\phi \psi_\phi(x)\chi_\phi(\xi), \tag{2}$$

where $\chi_n(\xi)$ and $\chi_q(\xi)$ are the bound and unbound relative motion eigenfunctions of the molecule. For brevity we use Greek letter indices to index both bound and unbound states. The unbound states are chosen to be discrete so that the unbound incident states have a finite numerical representation.

To obtain the probability that the two atoms combine to form a molecule, we take the atoms as being incident in an unbound molecular state $\chi_q(\xi)$ and calculate the probability that the reflected/transmitted state is a bound molecular state $\chi_n(\xi)$. We choose $\chi_n(\xi)$ and $\chi_q(\xi)$ to satisfy the relative motion Schrödinger equation with binding Hamiltonian H_b:

$$H_b\chi_\phi(\xi) = \left(-\frac{\hbar^2}{m}\frac{d^2}{d\xi^2} + V_0(\xi)\right)\chi_\phi(\xi) = e_\phi\chi_\phi(\xi), \tag{3}$$

where $e_\phi < 0$ for bound states and $e_\phi > 0$ for unbound states. Substituting Eq. (2) into Schrödinger's equation $H\Psi(x,\xi) = E\Psi(x,\xi)$, multiplying by $\chi^*_\mu(\xi)$ and integrating over ξ, we obtain

$$\left(\frac{d^2}{dx^2} + k_\mu^2\right)\psi_\mu(x) - \sum_\phi z_{\mu\phi}(x)\psi_\phi(x) = 0, \tag{4}$$

where the center-of-mass wave numbers are defined by

$$k_\mu^2 = \frac{4m}{\hbar^2}(E - e_\mu) \tag{5}$$

and the expressions for effective potentials $z_{\mu\nu}(x)$ are

$$z_{\mu\nu}(x) \equiv \frac{4m}{\hbar^2}\int_{-\infty}^{\infty}\left[V\left(x + \frac{1}{2}\xi\right) + V\left(x - \frac{1}{2}\xi\right)\right]\chi^*_\mu(\xi)\chi_\nu(\xi)d\xi. \tag{6}$$

The coefficients of reflection and transmission are extracted from Eq. (4) using the method of variable reflection and transmission amplitudes [13]. This method allows one to calculate the coefficients of transmission and reflection without having to calculate the wave function, thus reducing the amount of numerical computation substantially. A complete discussion on the application of this method is presented in Refs. [8,14]. The result is the following set of coupled, nonlinear, differential equations [14] which yield the reflection and transmission coefficients as boundary conditions:

$$\frac{d}{dy}R_{\mu\nu}(y) = -\sum_{\phi}\frac{1}{2ik_\phi}[e^{ik_\phi y}\delta_{\mu\phi} + e^{-ik_\phi y}R_{\mu\phi}(y)]\sum_{\gamma} z_{\phi\gamma}(y) \times[e^{ik_\gamma y}\delta_{\gamma\nu} + e^{-ik_\gamma y}R_{\gamma\nu}(y)], \tag{7}$$

$$\frac{d}{dy}T_{\mu\nu}(y) = -\sum_{\phi}\frac{1}{2ik_\phi}e^{-ik_\phi y}T_{\mu\phi}(y)\sum_{\gamma} z_{\phi\gamma}(y) \times[e^{ik_\gamma y}\delta_{\gamma\nu} + e^{-ik_\gamma y}R_{\gamma\nu}(y)], \tag{8}$$

subject to the boundary conditions

$$R_{\mu\nu}(y \to \infty) = 0, \quad R_{\mu\nu}(y \to -\infty) = R_{\mu\nu}, \tag{9}$$

$$T_{\mu\nu}(y \to \infty) = \delta_{\mu\nu}, \quad T_{\mu\nu}(y \to -\infty) = T_{\mu\nu}, \tag{10}$$

where $R_{\mu\nu}$ and $T_{\mu\nu}$ are the coefficients of reflection and transmission. The total probabilities of reflection and transmission in a bound molecular state $p_{\mathrm{RB}}^{(\nu)}$ and $p_{\mathrm{TB}}^{(\nu)}$ for the atoms incident in the state ν are

$$p_{\mathrm{RB}}^{(\nu)} = \sum_n \frac{k_n}{k_\nu}|R_{n\nu}|^2, \quad p_{\mathrm{TB}}^{(\nu)} = \sum_n \frac{k_n}{k_\nu}|T_{n\nu}|^2, \tag{11}$$

where n indexes all bound states. Similarly, the total probabilities of reflection and transmission in an unbound state for a molecule incident in the state ν are

$$p_{\mathrm{RU}}^{(\nu)} = \sum_q \frac{k_q}{k_\nu}|R_{q\nu}|^2, \quad p_{\mathrm{TU}}^{(\nu)} = \sum_q \frac{k_q}{k_\nu}|T_{q\nu}|^2, \tag{12}$$

where q indexes all unbound states. The total probabilities of reflection and transmission are

$$p_R^{(\nu)} = p_{\mathrm{RB}}^{(\nu)} + p_{\mathrm{RU}}^{(\nu)}, \quad p_T^{(\nu)} = p_{\mathrm{TB}}^{(\nu)} + p_{\mathrm{TU}}^{(\nu)}. \tag{13}$$

We also have $p_{\mathrm{RB}}^{(\nu)}+p_{\mathrm{TB}}^{(\nu)}+p_{\mathrm{RU}}^{(\nu)}+p_{\mathrm{TU}}^{(\nu)}=1$.

An alternative approach would be to use the time-dependent Schrödinger or Lippman-Schwinger equation and represent the molecule by a wave packet. One could then calculate the time evolution of the wave packet and obtain the probabilities of transmission and reflection. Jackson presented a review [15] explaining such an approach, using a Fourier grid for the packet to study chemical reaction dynamics, and noted that a physical and intuitive picture can be obtained using time-dependent methods. Use of wave packets and a treatment along these lines could apply to the problem studied here.

We choose the following binding potential:

$$V_0(\xi) = \begin{cases} V_2, & 0 < |\xi| < a, \\ -V_1, & a < |\xi| < b, \\ 0, & b < |\xi| < L, \\ \infty, & |\xi| > L. \end{cases} \tag{14}$$

This idealized potential captures the essential physics and reduces the amount of numerical calculation by giving simple, exact analytical expressions for the relative motion eigenfunctions $\chi_n(\xi)$ and $\chi_q(\xi)$, by solving Eq. (3).

Note that by the symmetry of x_1 and x_2 in the Hamiltonian, parity must be conserved. This implies that there is no even-odd coupling between the molecular states, i.e., the molecule cannot make a transition to a state of opposite parity. Therefore, without loss of generality, we discuss only the even relative motion states and we note that the results for a molecule incident in an odd state are similar to those given in this report. The even states are given in Ref. [14].

By varying the parameters in the binding potential we obtain the two (even) bound states in Ref. [14]. We index the even bound and unbound states in order of increasing energy, beginning with index "0," and incrementing by 2 (since the states are of even parity). For the case of two bound states we assign the indices "0" and "2" to the lower and higher energy bound states. The lowest energy unbound state is assigned the index "4," the next highest energy unbound state is indexed "6," and so on (odd states would be indexed by odd integers).

For eigenfunctions of even parity, the effective potential $z_{\mu\nu}(x)$, given by Eq. (6), reduces to

$$z_{\mu\nu}(x) = \frac{8m}{\hbar^2}\int_{-\infty}^{\infty} V\left(x + \frac{1}{2}\xi\right)\chi_\mu^*(\xi)\chi_\nu(\xi)d\xi. \tag{15}$$

For the external potential barrier we use a δ barrier of strength λ, given by $V(x \pm \frac{1}{2}\xi) = \lambda\delta(x \pm \frac{1}{2}\xi)$. This represents a δ barrier for the atoms at $x_j=0$, i.e., $V(x_j)=\lambda\delta(x_j)$ for $j=1,2$. We also use Gaussian barriers, as specified below.

The definitions of the dimensionless quantities we will use are

$$\tilde{x} \equiv \frac{x}{a}, \quad \tilde{k} \equiv ka, \quad \tilde{\lambda} \equiv \frac{\lambda}{aV_1}, \quad \tilde{z}_{\mu\nu} \equiv a^2 z_{\mu\nu}, \quad \tilde{L} \equiv \frac{L}{a},$$

$$f_n \equiv \frac{e_n}{V_1}, \quad f_q \equiv \frac{e_q}{V_1}, \quad N \equiv V_2/V_1, \quad g \equiv \sqrt{\frac{mV_1}{\hbar^2}}a. \tag{16}$$

The numerical values of the dimensionless parameters are chosen to be $g=15$, $N=5$, $b/a=1.3$, $\tilde{L}=10$, and $\tilde{\lambda}=0.01$. The calculated values of f_n are $f_0=-0.7226$ and $f_2=-0.02277$. The 15 values of f_q are $f_4=0.0006234$, $f_6=0.002487$, $f_8=0.005575$, $f_{10}=0.009865$, $f_{12}=0.01534$, $f_{14}=0.02198$, $f_{16}=0.02979$, $f_{18}=0.03874$, $f_{20}=0.04884$, $f_{22}=0.06008$, $f_{24}=0.07246$, $f_{26}=0.08598$, $f_{28}=0.1006$, $f_{30}=0.1164$, and f_{32}

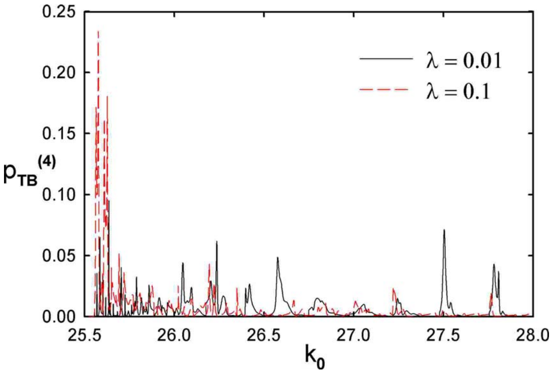

FIG. 1. (Color online) Shown is the probability $p_{\mathrm{TB}}^{(4)}$ that two atoms incident upon a single δ barrier will be transmitted in a molecular bound state, as a function of the (dimensionless) c.m. wave number k_0. Two different δ-barrier strengths are shown, $\lambda=0.01$ (solid curve) and $\lambda=0.1$ (dashed curve).

$=0.1334$. Reference [14] has details of these calculated values.

The values of the dimensionless parameters chosen above are not arbitrary, but rather have been chosen to be comparable to realistic systems. For example, taking m, V_1, and a to be on the order of an atomic mass unit, an electron volt, and the Bohr radius, respectively, one finds that g is of the same order as the value chosen above. Also, the dimensionless parameters have been chosen to give one deeply bound state and a second bound state that is also appreciably bound, i.e., the magnitude of its binding energy is not small.

III. RESULTS

We report three principal results. (1) Two atoms incident upon an external potential can combine to form a molecule that is either transmitted or reflected. (2) The probabilities p_{RB} and p_{TB} are maximized by adjusting the strength λ of the barrier. (3) The probabilities may be further increased if the atoms are incident upon multiple barriers; p_{TB} and p_{RB} can be as much as about 0.25 and 0.5, respectively, depending on the strength and separation of the individual barriers. The energies for the maximum probabilities are not obvious.

In Fig. 1 the probability of transmission in a bound molecular state $p_{\mathrm{TB}}^{(4)}$ is plotted as a function of the c.m. wave number k_0 for the atoms incident upon a single δ barrier. This plot represents the probability that the two atoms will tunnel through the barrier and emerge as a molecule on the other side. We consider the atoms incident in the lowest energy unbound molecular state, 4, and we note that higher energy incident states give similar results. By examining the probabilities of transition we find that $p_{\mathrm{TB}}^{(4)}$ is primarily due to transmission to the excited bound state 2 rather than the ground state 0. One might have expected transitions to the ground state to dominate. Nevertheless, this result agrees with previous studies involving transitions between molecular states in the process of tunneling [16]. The probability of transition from the incident state ν to the outgoing state n is given by

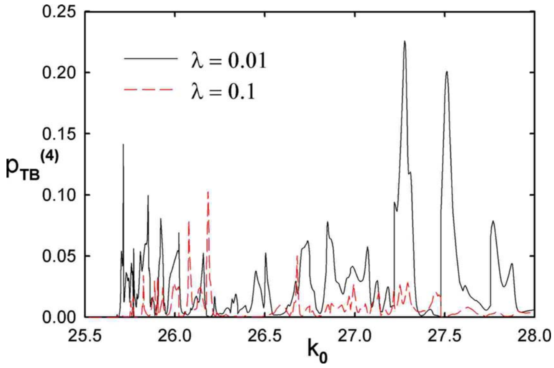

FIG. 2. (Color online) Shown is the probability $p_{\mathrm{TB}}^{(4)}$ that two atoms incident upon three δ barriers will be transmitted in a molecular bound state, as a function of the c.m. wave number k_0. The separation between the δ barriers is chosen to be $\langle\xi\rangle=1.174a$, i.e., the expectation value of the separation between the atoms in a bound, molecular state. Two different δ-barrier strengths are shown, $\lambda=0.01$ (solid curve) and $\lambda=0.1$ (dashed curve).

$$p_{\nu\to n}=\frac{k_n}{k_\nu}[|R_{n\nu}|^2+|T_{n\nu}|^2]. \qquad (17)$$

We find the probability of transmission in a bound molecular state to be quite sensitive to changes in barrier strength. The location and magnitudes of the local maxima vary significantly for different δ-barrier strengths. Moreover, the k_0 values for these maxima are not simply the values corresponding to the activation of higher energy unbound states. In Fig. 1 we present two different δ-barrier strengths $\lambda=0.01$ (solid curve) and $\lambda=0.1$ (dashed curve). Observe that the probability of transmission in a molecular bound state is much larger for the stronger δ barrier at low incident energies and opposite so for larger incident energies. Indeed, for $k_0\approx 25.6$, $p_{\mathrm{TB}}^{(4)}$ is almost 0.25 for the stronger barrier whereas $p_{\mathrm{TB}}^{(4)}$ is no more than 0.1 for the weaker barrier. One might expect the probability of reflection to be larger for the stronger barrier at low incident energies. Intriguingly, this is not the case and reasons for this are not clear due to the complexity of the coupling between the amplitudes for the four probabilities $p_{\mathrm{RB}}^{(4)}$, $p_{\mathrm{TB}}^{(4)}$, $p_{\mathrm{RU}}^{(4)}$, and $p_{\mathrm{TU}}^{(4)}$; see Eqs. (7) and (8). For $k_0>26.5$ we find that $p_{\mathrm{TB}}^{(4)}$ is generally much larger in magnitude for the weaker δ barrier than for the stronger δ barrier.

Next we investigate the probability of transmission in a bound molecular state for the atoms incident upon multiple δ barriers. The locations and magnitudes of the local maxima change unpredictably with the inclusion of additional barriers. This is because the local maxima are the result of interference between incident and reflected waves which depend sensitively upon the number and separation of the δ barriers. We find that three δ barriers give the largest magnitudes of $p_{\mathrm{TB}}^{(4)}$ while two, four, five,…, δ barriers give qualitatively similar but generally smaller probabilities of forming a molecule. Figure 2 shows $p_{\mathrm{TB}}^{(4)}$ versus k_0 for the atoms incident upon three δ barriers. It is found that the probability of forming a molecule is maximized when the separation between the external barriers is equal to the expectation value of the separation between the two atoms in a bound state,

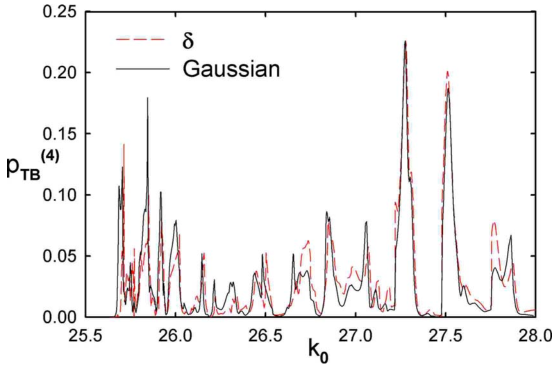

FIG. 3. (Color online) $p_{\mathrm{TB}}^{(4)}$ vs the c.m. wave number k_0 comparing three δ barriers to three Gaussian barriers, all with $\tilde{\sigma}=0.03$. The areas of the Gaussian barriers are all chosen to be equal to the strengths of the δ barriers, i.e., $A=\tilde{\lambda}=0.01$. The δ barriers are clearly a good approximation of the Gaussian barriers. The separation between the barriers is $\langle\xi\rangle$.

$\langle\xi\rangle=1.174a$. The external potential is thus chosen to be $V(x)=\delta(x+\langle\xi\rangle)+\delta(x)+\delta(x-\langle\xi\rangle)$.

Note that the δ barriers are arranged symmetrically about the origin. It is also found that barrier separations of half integral multiples of $\langle\xi\rangle$, i.e., $\frac{1}{2}\langle\xi\rangle$, $\langle\xi\rangle$, $\frac{3}{2}\langle\xi\rangle,\ldots$, give optimal probabilities of forming a molecule. Deviations from these barrier separations of half integral multiples of $\langle\xi\rangle$ result in significant reductions in $p_{\mathrm{TB}}^{(4)}$, especially for larger numbers of δ barriers (due to a compounding effect). For multiple barriers we again find that $p_{\mathrm{TB}}^{(4)}$ is primarily due to the atoms tunneling in the excited bound state while the probability of tunneling in the ground state is much smaller.

Figure 3 compares the results for three δ barriers to those for three Gaussian barriers given by $V(x)=\frac{A}{\sigma\sqrt{2\pi}}[\exp(-\frac{(x+\langle\xi\rangle)^2}{2\sigma^2})+\exp(-\frac{x^2}{2\sigma^2})+\exp(-\frac{(x-\langle\xi\rangle)^2}{2\sigma^2})]$. The Gaussian barriers chosen are modestly wide with $\tilde{\sigma}\equiv\sigma/a=0.03$ in order to show a deviation from the results of the δ barriers. We note that for $\tilde{\sigma}<0.03$ the results for the Gaussian barriers and the δ barriers are almost identical. Hence, the δ barriers serve as an excellent approximation to more realistic, spatially narrow barriers.

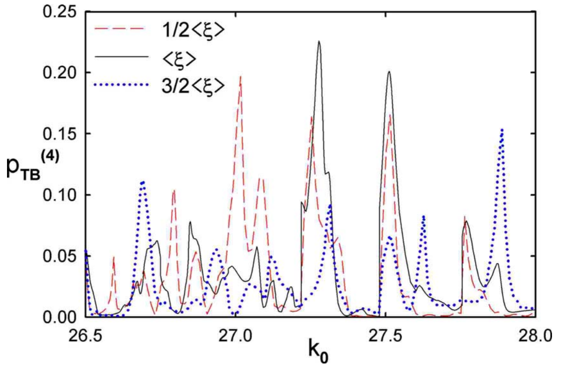

FIG. 4. (Color online) Shown are the probabilities $p_{\mathrm{TB}}^{(4)}$ that two atoms incident upon three regularly spaced δ barriers will be transmitted in a molecular bound state, as a function of the c.m. wave number k_0. Three different spacings are shown: $\frac{1}{2}\langle\xi\rangle$ (dashed curve), $\langle\xi\rangle$ (solid curve), and $\frac{3}{2}\langle\xi\rangle$ (dotted curve), where $\langle\xi\rangle$ is the expectation value of the separation between atoms in a bound molecular state.

Figure 4 shows the manner in which the probability of transmission in a bound state depends upon the separation between three δ barriers. The dashed curve is for barriers separated by half the expectation value of the separation between atoms in a bound molecular state, i.e., $\frac{1}{2}\langle\xi\rangle$. The solid curve is for barriers separated by $\langle\xi\rangle$ and the dotted curve is for $\frac{3}{2}\langle\xi\rangle$. In all cases the probabilities have peaks with relatively large values. Separation by $\langle\xi\rangle$ results in two high peaks in the probability, approximately 0.20 and 0.23. Spacing the barriers by $\frac{1}{2}\langle\xi\rangle$ gives five peaks with probabilities in the approximate range 0.10 to 0.20. Spacing by $\frac{3}{2}\langle\xi\rangle$ gives several peaks but only two with peak values exceeding 0.10. These separation distances are multiples of half the expectation value of the length of the molecule. For larger multiples, the peak probability values decrease from those in Fig. 4.

IV. SUMMARY AND FURTHER WORK

The results in this paper can be extended from one dimension to three dimensions. Consideration of the rotational and vibrational modes of a molecule tunneling in three dimensions with only bound states was investigated in Ref. [9]. The freedom associated with the rotational modes allows for orientations of the axis of the molecule which are not perpendicular to the barrier. Many of the characteristics observed in the one dimensional study in this paper will also be found in a three-dimensional investigation. Additional considerations of the already complicated internal structure of a diatomic molecule could be investigated, including, for example, the spins of the atoms [5]. It has been demonstrated that it is possible to align molecules in a molecular beam using laser pulses, optical fields, dc electric fields, and inhomogeneous electric fields [17]. Such methods might permit a situation in which the results of this paper could be applied in an experimental setting. The relationship between scattering and tunneling problems provides additional examples of applications in molecular physics. The scattering of H_2 from Cu(001) [18] and NO from diamond (110) [19] have been shown to produce energy resonances analogous to those presented in this one-dimensional model. Furthermore, it has been shown that the scattering distributions depend on the incoming and outgoing states (i.e., vibrational and rotational excitations) similar to the internal excitations included in our model.

We found that two atoms incident upon an external potential can combine to form a molecule. The probability of forming a molecule showed several local maxima at various incident energies. The magnitudes and locations of these maxima were found to be sensitive to changes in the external potential. The probability of forming a molecule can be enhanced by having the two atoms incident upon multiple potential barriers. We found that three delta or Gaussian barriers give the largest probability of forming a molecule. The optimal separation between multiple barriers was found to be half integral multiples of the expectation value of the separation of the two atoms in a bound state. This is as one would expect. Further enhancements in the probability resulted by varying the strengths of the barriers. The probability of forming a molecule from two atoms incident upon a potential barrier was shown to be as high as 0.5 for reflection and 0.25 for transmission. The results for δ barriers were in excellent agreement with the results for spatially narrow Gaussian barriers.

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