Beyond universality: Parametrizing ultracold complex-mediated reactions using statistical assumptions

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We have calculated accurate quantum reactive and elastic cross sections for the prototypical barrierless reaction $D^+ + H_2(v = 0, j = 0)$ using a modified hyperspherical scattering method. The considered kinetic energy ranges from the ultracold to the Langevin regimes. A reaction rate coefficient practically constant in no less than eight orders of magnitude is obtained. The availability of accurate results for this system allows one to test the quantum theory by Jachymski *et al.* [K. Jachymski, M. Krych, P. S. Julienne, and Z. Idziaszek, Phys. Rev. Lett. **110**, 213202 (2013)] in a nonuniversal case. The short-range reaction probability is rationalized using statistical model assumptions and related to a statistical factor. This provides a means to estimate one of the parameters that characterizes ultracold processes from first principles.

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The increasing availability of cold and ultracold samples of atoms and molecules has sprung great interest in chemical reactions at very low temperatures [1–5]. Although new experimental approaches [5] appear highly promising, advances in the field are hampered by technical problems in producing most molecules at low temperatures and high enough densities. In contrast to neutral species, ions can be easily trapped and cooled. The technology of Coulomb crystals in radio-frequency ion traps [6] and the possibility of combining them with traps for neutrals or with slow molecular beams [7,8] promise great progress in the analysis of ion-neutral reactions in the near future.

Theoretical simulations employing standard ab initio approaches are not feasible for most of the systems thus far considered. For heavy systems (more convenient experimentally) there are no potential energy surfaces (PESs) accurate enough to describe processes near thresholds. Additionally, most of exact dynamical treatments face insurmountable problems in such regimes. However, in contrast to short-range (SR) chemical interactions, those occurring at long range (LR) can be more easily calculated. Moreover, theoretical approaches based only on the knowledge of the LR part of the PES have been able to describe recent experimental findings nearly quantitatively [1,9]. Indeed, processes at very low collision energies favor LR interactions, leading to the idea of *universality* in extreme cases [10]: the result of the collision depends exclusively on the LR behavior and not on the details of the PES. In this regard, recently proposed LR parametrization procedures [9,11–13] are very appealing.

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Fitting experimental data, these models are able to predict nonmeasured values providing some insight into the underlying interactions. In particular, the approach by Jachymski *et al.*, based on multichannel quantum-defect theory (MQDT), provides analytical expressions which can be easily compared with experimental data [9,14,15]. The model has been recently applied to a variety of systems [9,16,17]. In particular, for the Penning ionization of Ar by He(³S) [5], the rate coefficients have been fitted in a wide range of collision energies using only two parameters [9]. However, the parameters of the model are phenomenological and they had not been determined before from first principles.

In this work, we present accurate calculations for the reactive collision $D^+ + H_2(v = 0, j = 0)$ using the hyperspherical reactive scattering method [18]. We consider collision energies that range from the ultracold regime, where only one partial wave is open, to the Langevin regime where many of them contribute. These calculations allow us to test the model by Jachymsky *et al.* [9,14] by comparison with accurate theoretical results in a realistic atom + diatom system, providing a way to estimate one of the parameters using simple statistical model assumptions, which do not require performing any quantum reactive scattering calculation.

The H⁺ + H₂ system is the prototype of ion-molecule reactions, which are usually nearly barrierless and exhibit large cross sections due to their LR, $\alpha - C_n/R^n$, n = 4, potentials. At energies below ≈ 1.7 eV, the proton exchange is the only reactive channel, and the process can be described on the ground adiabatic PES [19–22]. Since the PES is characterized by a deep well or complex (≈ 4.5 eV), as illustrated in Fig. 1, rigorous statistical models [23,24] have been applied to this reaction and isotopic variants in the low and thermal energy regimes [20,24–27] in good agreement with accurate

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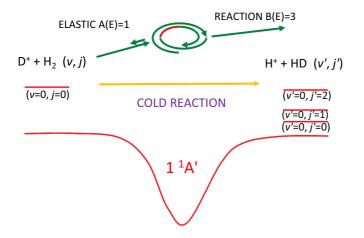


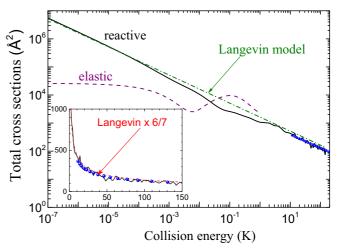
FIG. 1. (Color online) Sketch of the intrinsic reaction path and the rovibrational states involved in the reaction at the studied energies. A(E) and B(E) are the number of incoming and outgoing channels, respectively (1 and 3 for J = 0).

calculations. Specifically, the $D^+ + H_2 \rightarrow H^+ + HD$ reaction features a small exoergicity (difference of zero-point energies). Experiments to determine state-specific rate coefficients at energies as low as 12 K [28] have been carried out, and lower temperatures are expected to be feasible soon [29].

The deep ultracold regime, governed by Wigner laws [30], is described in terms of the scattering length. The latter largely varies with slight changes of the interaction potential. Due to inaccuracies in the state-of-the-art electronic calculations, only for very particular atom + atom systems [31–33] has it been possible to reproduce the experimental scattering length theoretically. As atom + diatom systems are even more complicated, it is difficult to assess the accuracy of calculations for ultracold energies. In the spirit of the work by Gribakin *et al.* [34], we can consider our study as an effort to determine a "characteristic" scattering length. Besides, assuming that the interaction of the system is reasonably described by the current PES, we can use our results to test recent methodologies, like the approach in Refs. [9,14].

In the (ultra)cold regime both accurate descriptions of the LR interactions and dynamical propagations up to very large distances are two strict requirements. The PES by Velilla *et al.* [35], which includes the LR interactions in the functional form, satisfies the first requirement. The dominant contributions involve the charge quadrupole, $\propto -R^{-3}$, and the charge-induced dipole, $\propto -R^{-4}$, interactions. However, only the latter contributes to collisions in j = 0.¹ The second requirement is fully satisfied by the hyperspherical quantum reactive scattering method developed by Launay *et al.* [18,36], recently modified to allow the inclusion of LR interactions [37,38]. These modifications are used here for the first time allowing the propagations up to 10^5-10^6 a.u. which are required for a n = 4 potential.

The reaction cross section, $\sigma_r(E)$, in the 10^{-7} –150 K energy range is plotted in Fig. 2 and compared with the Langevin



PHYSICAL REVIEW A 91, 030701(R) (2015)

FIG. 2. (Color online) Calculated reaction and elastic total cross section for the collision $D^+ + H_2 (v = 0, j = 0)$ compared with the Langevin prediction. The inset shows the comparison of the reaction cross sections with the experimental data [28] and the result of the statistically corrected Langevin model.

model (LM) expression for the cross section, $\sigma_L(E) = 2\pi (C_4/E)^{1/2}$. The LM is commonly used to rationalize collisions in the regime where many partial waves are open. We have determined the value $C_4 = 2.71$ a.u. using the effective potential as a function of R which results from averaging the PES $V(R, r, \theta)$ over r and θ Jacobi coordinates, with the (v = 0, j = 0) probability distribution. The LM implies here that once the centrifugal barrier is overcome, the system is captured in the complex which subsequently decomposes into the H⁺ + HD arrangement channel with unit probability. The LM regime can be associated with the high energy part of the plot (above 1 K, with five partial waves opened). The calculated cross sections are found smaller than the LM prediction in this energy range. Indeed, only a fraction of the complexes decompose into the products.

We can improve the LM using statistical model arguments, which are being revisited in the field of cold collisions [39,40]. In complex mediated reactions, the statistical ansatz [23,24], $P_r^J(E) \approx P_{\text{capt}}^J(E) \times P_{\rightarrow \text{prod}}(E)$, can be applied, where $P_r^J(E)$ is the reaction probability for a given initial rovibrational state and total angular momentum, J (orbital, l, plus rotational, j), $P_{\text{capt}}^{J}(E)$ is the probability for the reagents to be captured in the complex, and $P_{\rightarrow \text{prod}}(E)$ is the statistical factor, i.e., the probability of emerging into the product arrangement channel when the complex decomposes. If there is a complete randomization of the energy in the complex, the statistical factor will be independent of the initial state of the reagents, only subject to conservation of energy, J, and parity. Roughly speaking, the fraction of complexes which decompose into the reactants or products is proportional to the respective number of scattering channels energetically available, denoted with A(E) and B(E), respectively, considering all of them as equiprobable. Accordingly, the statistical factor can be approximated by $P_{\rightarrow \text{prod}}(E) = B(E)/[A(E) + B(E)]$. At the considered energies, only three HD rovibrational states are open, as shown in Fig. 1, and for $J \ge 2$ we find that A(E) = 1and B(E) = 6, and $P_{\rightarrow \text{prod}} = 6/7 (\approx 86\%)$. For J = 0 and

¹The integral $\langle j = 0 | P_2 | j = 0 \rangle$, where $P_2(\cos \theta)$ is the second order Legendre polynomial, is null, and the contributions from $\sim R^{-3}$ and anisotropic polarization terms vanish [35].

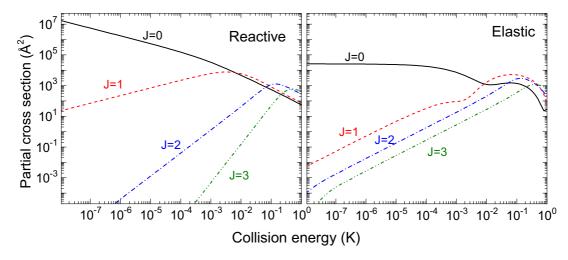


FIG. 3. (Color online) Reaction and elastic partial cross sections at the indicated J (= l) values for the D⁺ + H₂ (v = 0, j = 0) collision in the cold and ultracold regimes.

J = 1 the statistical factors are 3/4 (75%) and 5/6 (\approx 83%), respectively. If the collision energy is high enough for many partial waves to contribute but low enough for the total number of channels to stay the same, $P_{\rightarrow \text{prod}}(E) \approx 6/7$, and

$$\sigma_{\rm r}(E) \approx P_{\rightarrow \,\rm prod} \,\sigma_{\rm L}(E).$$
 (1)

Therefore, 6/7 appears naturally as a statistical factor to correct the LM expression. When the number of product channels is large enough, $B \gg A$, then $P_{\rightarrow \text{prod}} \approx 1$ and the result is $\sigma_L(E)$. More accurate statistical implementations, which evaluate A(E) and B(E) as capture probabilities [23,24], lead to similar conclusions.

The inset of Fig. 2 compares the calculated reaction cross section with the experimental data from Ref. [28]. The corrected LM result is also shown and found to be in very good agreement with both the experiment and the present calculations. The similarity of the theoretical results with the experiment is remarkable considering that the latter was performed with n-H₂ [21] and lends credence to the predictive power of QM calculations on state-of-the-art PESs for atom + diatom systems *ab initio* in the Langevin regime.

The cross sections at much lower kinetic energies are also shown in Fig. 2. In the zero-energy limit, Wigner threshold laws [30,41] state that the elastic and the total-loss (inelastic + reaction) cross sections associated with each partial wave, l, vary $\sim E^{2l}$ and $\sim E^{l-1/2}$, respectively. However, for a potential with n = 4, the threshold law for elastic scattering becomes $\sim E$ for any l > 0 [30,41,42]. The ultracold cross sections, shown in Fig. 3 for the four lowest partial waves, comply with these laws (there are no open inelastic channels). The limiting behaviors for l = 0 are reflected in the total reaction ($\sim E^{-1/2}$) and total elastic (constant) cross sections in the lowest energy region of Fig. 2, where only the *s* wave is open.

For n = 4 the energy dependence of the LM coincides with the Wigner threshold law ($\sim E^{-1/2}$). Remarkably, the absolute values of accurate and LM cross sections in the ultracold limit are nearly the same, $\sigma_r \approx 1.07\sigma_L(E)$. Therefore, the reaction rate coefficient (not shown) is practically constant in no less than eight orders of magnitude, and small variations can be further smoothed out with the Boltzmann averaging. In what follows, we will try to rationalize this classical Langevin behavior in the ultracold regime.

Very recently, quantal versions of the LM have been proposed [14,43] under the assumption that all the flux that reaches the SR region leads to reaction. In the n = 4 case, these *universal* models conclude that the zero-energy limit of σ_r is given by $2\sigma_{\rm L}(E)$, and not by $\sigma_{\rm L}(E)$ as we have approximately obtained. Therefore our system is not universal. The formalism in Ref. [9] is able to deal with systems where the short-range reaction probability, Pre, is <1. It provides expressions for the complex (energy-dependent) scattering length $\tilde{a}_l(k) =$ $\alpha_l(k) - i\beta_l(k)$ in terms of the MQDT functions (where k is the relative wave number). This allows us to parametrize $\tilde{a}_l(k)$ using two real parameters, y and s, together with the mean scattering length [34], $\bar{a} = (2\mu C_4)^{1/2}/\hbar ~(\approx 99.7 a_0)$ in this case). Specifically, the dimensionless parameter $0 \leq$ $y \leq 1$ characterizes the flux that is lost from the incoming channel at SR, according to $P^{re} = 4y/(1+y)^2$. The Langevin assumption or *universal case* corresponds to y = 1. The dimensionless scattering length $s = tan(\phi)$ is related to an entrance channel phase ϕ [9,14,42].

In terms of these parameters, the small k behavior of the real and imaginary parts of the complex scattering length for the lowest partial waves (l = 0-3) is given by²

$$\alpha_0(k) \to \bar{a} \frac{s(1-y^2)}{1+s^2 y^2}, \quad \beta_0(k) \to \frac{y(1+s^2)\bar{a}}{1+s^2 y^2},$$
(2)

$$\alpha_2(k) \to -k\bar{a}^2 \frac{\pi}{105}, \quad \beta_2(k) \to \frac{y(1+s^2)k^4\bar{a}^5}{2025(1+s^2y^2)}, \quad (4)$$

$$\alpha_3(k) \to -k\bar{a}^2 \frac{\pi}{315}, \quad \beta_3(k) \to \frac{y(1+s^2)k^6\bar{a}^7}{2\,480\,625(s^2+y^2)}.$$
(5)

²Equations (2) and (3) were kindly provided by the authors of Ref. [9]; Eqs. (4) and (5) were deduced by the authors of this work following Refs. [9,42].

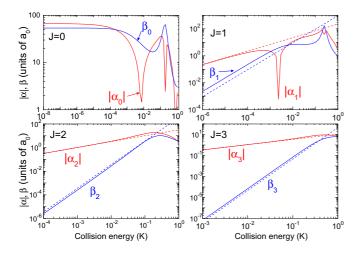


FIG. 4. (Color online) Real and imaginary parts of the scattering length, $\tilde{a}_l(k) = \alpha_l(k) - i\beta_l(k)$, obtained in the calculations for the four lowest partial waves. The absolute value of α , mostly negative, is plotted. The values for J = 1, 2, and 3 (in continuous lines) are compared with the predictions from the model in Ref. [9] (in dashed lines), calculated using Eqs. (3)–(5), assuming for *s* and *y* the same values that have been obtained for J = 0.

Our calculations yield the *S* matrix as a function of the energy for each total angular momentum *J* and hence *l* (for j = 0), which allows us to calculate directly $\tilde{a}_l(k)$ using the elastic element of the *S* matrix [44].

Figure 4 depicts the energy dependence of α and β for J = 0-3. The limiting behaviors are in perfect agreement with the threshold laws and the power of the dependence on k in Eqs. (2)–(5).

To extract the model parameters *s* and *y* from the scattering results, let us consider first the case l = 0. Using the values α_0 and β_0 at the lowest energy given by our calculations and solving Eqs. (2) for *y* and *s*, we obtain y(l = 0) = 0.35 and s(l = 0) = -0.82, which leads to $P^{\text{re}}(l = 0) = 77\%$. The parametrization for higher values of *l* is not straightforward. The real part, $\alpha_l(k)$, is independent on s(l) and y(l), and with the sole expression of $\beta_l(k)$ it is not possible to solve for the values of the two parameters.

Analogous to the procedure of Ref. [9], assuming that y and s do not depend on l, we can introduce y(l = 0) and s(l = 0) in Eqs. (3)–(5) and compare the resulting values of β_l with those obtained in the scattering calculations. The ratios of the calculated and parametrized values of β_l are 0.4, 1.4, and 0.7 for l = 1, l = 2, and l = 3, respectively. The agreement can be considered good on average, taken into account the oscillations of this ratio about 1. As for the real parts α_l , given by Eqs. (3)–(5), they can be directly compared with our

PHYSICAL REVIEW A 91, 030701(R) (2015)

scattering results. The agreement (within 1%) is very good, which can be deemed as a test of the theory and serves to ensure the convergence of the scattering calculations. These expressions depend only on \bar{a} (not on *s* or *y*) and they can be considered as really universal.

According to Ref. [9], when the dependence of P^{re} with the energy and l is weak, the expression

$$\sigma_{\rm r}(E) \approx P^{\rm re} \sigma_{\rm L}(E) \tag{6}$$

is valid in the Langevin regime, P^{re} being the same value which governs the ultracold behavior. The fact that $\sigma_{\text{r}}(E)/\sigma_{\text{L}}(E)$ has an average value of 0.78 in the range 1–150 K, very close to the $P^{\text{re}}(l=0) = 0.77$ obtained at ultracold energies, indicates a weak dependence of P^{re} with l and energy and hence the validity of the assumption of constant parameters made above.

Therefore, Eq. (1), obtained from the statistical hypothesis, and Eq. (6), from Ref. [9], are both valid in the Langevin regime. This leads to the equivalence of the fraction of captured flux that reacts, $P^{\rm re}$, and the fraction of formed complexes which decompose to give the products, $P_{\rightarrow \text{prod}}$, and thus a way to estimate the P^{re} . In fact, 6/7 ($\approx 86\%$) is a rough estimate of $P^{\rm re} = 77\%$, and only requires counting states. A more accurate estimate can be obtained using the *ab initio* QM values of $\sigma_{\rm r}(E)$ in the Langevin region to calculate $\sigma_{\rm r}(E)/\sigma_{\rm L}(E)$. This second way to estimate $P^{\rm re}$ does not require the system to behave statistically and is more general. Finally, the fact that our calculations are expected to be quantitative in the Langevin regime leads to an interesting conclusion: we have found a way to estimate P^{re} , and through it y, one of the parameters which characterize the experimental ultracold behavior, while working at much higher energies where state-of-the-art ab initio reaction dynamics is quantitative and require less demanding QM calculations.

In summary, accurate scattering calculations have allowed us to test the quantum theory by Jachymsky *et al.* [9,14,15]. On average, the same P^{re} describes the ultracold and the Langevin regimes. We have found a link between P^{re} and the statistical factor of the statistical approach to reactions [23]. Apart from physical insight, our analysis provides ways to estimate the parameter *y*, which characterizes the experimental ultracold behavior, using *ab initio* reaction dynamics at much higher energies, where such methodology is assumed to be quantitative.

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