ON THE VALIDITY OF COLLIDER-MASS SCALING FOR MOLECULAR ROTATIONAL EXCITATION

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ABSTRACT

Rate coefficients for collisional processes such as rotational and vibrational excitation are essential inputs in many astrophysical models. When rate coefficients are unknown, they are often estimated using known values from other systems. The most common example is to use He-collider rate coefficients to estimate values for other colliders, typically H_2, using scaling arguments based on the reduced mass of the collision system. This procedure is often justified by the assumption that the inelastic cross section is independent of the collider. Here we explore the validity of this approach focusing on rotational inelastic transitions for collisions of H, para-H_2, ^{3}He, and ^{4}He with CO in its vibrational ground state. We compare rate coefficients obtained via explicit calculations to those deduced by standard reduced-mass scaling. Not surprisingly, inelastic cross sections and rate coefficients are found to depend sensitively on both the reduced mass and the interaction potential energy surface. We demonstrate that standard reduced-mass scaling is not valid on physical and mathematical grounds, and as a consequence, the common approach of multiplying a rate coefficient for a molecule-He collision system by the constant factor of ∼1.4 to estimate the rate coefficient for para-H_2 collisions is deemed unreliable. Furthermore, we test an alternative analytic scaling approach based on the strength of the interaction potential and the reduced mass of the collision systems. Any scaling approach, however, may be problematic when low-energy resonances are present; explicit calculations or measurements of rate coefficients are to be preferred.

Key words: molecular data – molecular processes – scattering

Online-only material: color figures

1. INTRODUCTION

The modeling of astrophysical observations requires a large variety of fundamental data. In cool, low-density molecular regions, collisional excitation/de-excitation rate coefficients are one class of data needed for such models. In many cases, however, the (de)excitation rate coefficients for systems of astrophysical interest are not available. While many molecular collisional calculations have been performed using He as a collider due to its relative ease of computation, the dominant neutral species in many astrophysical environments is H_2 or H. A common practice to obtain estimates for these unknown rates is to approximate them from known rate coefficients from other collision systems, He colliders, for example, by a reduced-mass scaling relation (e.g., Schöier et al. 2005; Van der Tak 2011). This procedure has generally been ascribed to Green et al. (1978), who predicted excitation rates of H_2 to be about 30% higher than He rates in collisions with H_2CO, and later computed broadening cross sections from line-width parameters and predicted state-to-state excitation rate coefficients for CO–H_2O collisions (Green 1993). These predictions were compared to theoretical He–CO rate coefficients and it was found that rate coefficients for excitation by water were related to those by He through the square root of the ratio of the systems' reduced masses. Although the experimental data limited the applicability of the predictions to room temperature and above, this "standard" reduced-mass scaling relation has been used extensively for lower temperatures and for other collisional parameters, e.g., inelastic rate coefficients.

Recently, the accuracy of the standard reduced-mass scaling approach has been tested for SiS (Lique et al. 2008) and SO_2 (Cernicharo et al. 2011) excitation and found to be inaccurate (see also Roueff & Lique 2013; Van der Tak 2011). In this paper,

we revisit reduced-mass scaling and investigate how the reduced mass μ and interaction potential energy surface (PES) impact explicitly computed rate coefficients. Due to the wealth of data on CO, and its importance in astrophysics, it is adopted here as a test molecule and used to explore more physically reasonable analytical and scaling approximations to estimate unknown rate coefficients.

2. METHOD

Quantum mechanical close-coupling calculations were performed using the nonreactive scattering program MOLSCAT (Hutson & Green 1994) with the modern PESs of Shepler et al. (2007), Jankowski & Szalewicz (2005), and Heijmen et al. (1997) for H–, H_2–, and He–CO, respectively. The scattering cross sections were computed for kinetic energies between 10^{-6} and 10^3 cm^{-1} within the rigid-rotor approximation with CO in its vibrational ground state and H_2 it is rovibrational ground state, i.e., para-H_2. Only para-H_2 ($j = 0$) is considered in our study; ortho-H_2 and para-$H_2(j > 0)$ rate coefficients obtained via any mass scaling approach from He data are unlikely to be accurate due to the lack of spherical symmetry of the internally excited molecule. De-excitation rate coefficients as a function of temperature T were obtained by thermally averaging the cross sections over a Maxwellian kinetic energy distribution given by

$$k_{j \to j'}(T) = \left(\frac{8k_B T}{\pi \mu}\right)^{1/2} \frac{1}{(k_B T)^2} \int_0^\infty \sigma_{j \to j'}(E_k) \\ \times \exp(-E_k/k_B T)E_k dE_k, \quad (1)$$

where $\sigma_{j \to j'}(E_k)$ is the state-to-state rotationally inelastic cross section, E_k the center of mass kinetic energy, $\mu = m_X m_{CO}/(m_X + m_{CO})$ the reduced mass of the X–CO system for
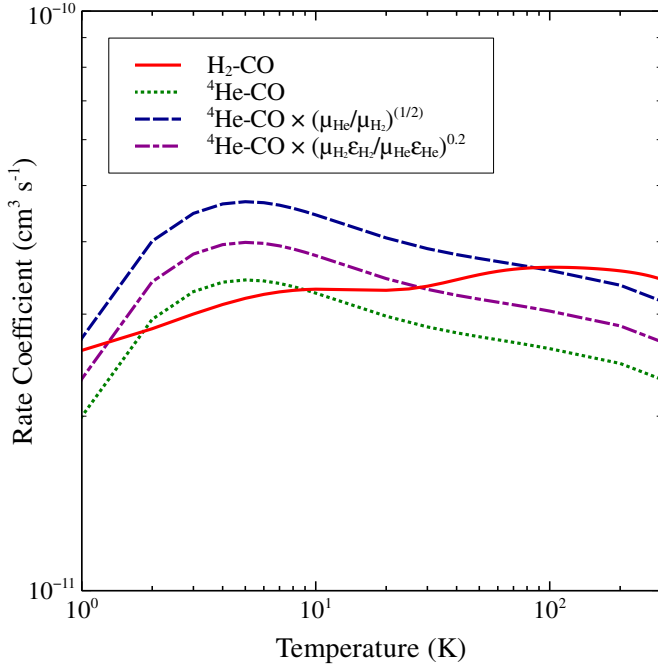
Figure 1. Rate coefficients for the de-excitation of CO($j = 1$) with the colliders He and para-H$_2$ and estimated values for H$_2$ via standard reduced-mass scaling and reduced-potential scaling.

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collider X, k_B the Boltzmann constant, and j the CO rotational quantum number.

3. RESULTS AND DISCUSSION

The main assumption of standard reduced-mass scaling is the statement that the cross section as a function of E_k is independent of the collider. It is then argued that the rate coefficients between He–CO and H$_2$–CO, for example, scale as the square root of the ratio of reduced masses according to

$$k_{j \to j'}^{H_2}(T) = \left(\frac{\mu_{He}}{\mu_{H_2}}\right)^{1/2} k_{j \to j'}^{He}(T), \qquad (2)$$

as deduced from the prefactor of Equation (1). Here $k_{j \to j'}^{X}(T)$ is the state-to-state rate coefficient for collider X, μ_X is the reduced mass for the X–CO system, and a prefactor of $\sim$1.4 is obtained in this case. As an illustration, Figure 1 displays the calculated rate coefficients for the de-excitation of CO($j = 1$) with the colliders He and H$_2$. The estimated H$_2$ rate coefficients using the standard reduced-mass scaling relation, Equation (2), are seen to deviate significantly from the explicitly computed values, especially for $T \lesssim 100$ K.

Although the standard reduced-mass scaling relation has been widely adopted (e.g., Schöier et al. 2005; Adande et al. 2013; Matsuura et al. 2014), it actually assumes not that the cross section $\sigma(E_k)$, but that the integral in Equation (1), is independent of the collider. However, this assumption is not generally valid because (a) the cross section depends on the adopted PES and (b) the kinetic energy depends on μ. In cases where Equation (2) has produced reasonable estimates, it may have been the result of fortuitous cancelation of the effects due to points (a) and (b).

To explicitly illustrate the failings of Equation (2) and to explore other more physically motivated approaches, we investigate three tracks: (1) the behavior of inelastic cross sections as a function of system parameters, (2) prediction of $k_{j \to j'}(T)$ adopting analytical relations for the cross section, and (3) an alternative scaling approach based on the well depth of the PES. To test the dependence of the cross section on μ and the PES, a series of calculations were performed for the collider masses H, H$_2$, ^{3}He, and ^{4}He with CO on each of the H–CO, H$_2$–CO, and He–CO PESs with some examples shown in Figure 2. In Figure 2(a), the cross sections, which were all calculated on the He–CO PES, are seen to depend significantly on the adopted μ. In addition to changes in the cross section slopes and magnitudes, the positions of quasibound resonances vary, especially for H$_2$ and He compared to H. Figure 2(b) shows the results of calculations using the ^{4}He–CO reduced mass on the three different PESs. Results for the other three masses (not shown) were likewise found to be sensitive to the PES. As expected, the cross section does indeed depend on both the PES and μ, and the assumptions implicit in Equation (2) are not valid.

As a yet further illustration, Equation (1) can be rewritten with the cross section given in terms of the relative velocity v of the collision system (Flower 1990):

$$k_{j \to j'}(T) = \left(\frac{2}{\pi}\right)^{1/2} \left(\frac{\mu}{k_B T}\right)^{3/2} \int_0^\infty \sigma_{j \to j'}(v)$$
$$\times \exp(-\mu v^2 / 2 k_B T) v^3 dv. \qquad (3)$$

This leaves μ in both the exponential Boltzmann term in the integral and in the prefactor and shows that the original arguments justifying standard reduced-mass scaling should be reconsidered (compare to Equation (1)). To gain additional insight and to explore an alternate scaling approach, assume the cross section to have the analytical form

$$\sigma_{j \to j'}(v) = B v^a, \qquad (4)$$

where B is an (undetermined) constant and a is some power. This leads to rate coefficients of the form (Stancil et al. 1998)

$$k(T) = A(a) B(T/\mu)^b, \qquad (5)$$

where $b = (1/2)(a + 1)$ and A is a function of a, both deduced from the Gaussian integral in Equation (3). This result is exact, given the assumption of Equation (4), and applicable to all collision systems. Therefore, if $\sigma(v)$ is assumed to be independent of the collider as originally supposed, Equation (2) is corrected by replacing the square root with the exponent b yielding

$$k_{j \to j'}^{Z}(T) = \left(\frac{\mu_Y}{\mu_Z}\right)^b k_{j \to j'}^{Y}(T). \qquad (6)$$

The scaling equation is now general for any two colliders Y and Z and any dependence of cross section on energy. Only for a constant cross section will b equal 1/2.

Figure 3(a) displays the cross section dependence for ^{3}He–CO and ^{4}He–CO, but plotted as a function of the kinetic energy divided by μ, which is proportional to v^2. Above $\sim$0.1 cm^{-1} u^{-1}, the cross section is relatively independent of μ, a concept well known in ion–atom collisions (e.g., Stancil & Zygelman 1995). Figure 3(b) gives a related plot where He and para-H$_2$ colliders give qualitatively similar behavior, with the background cross sections of each falling-off with a $1/v$ dependence for $E \gtrsim$ 2 cm^{-1} u^{-1}. Combining these observations with Equations (5) and (6), the resulting rate coefficients, neglecting the resonances,
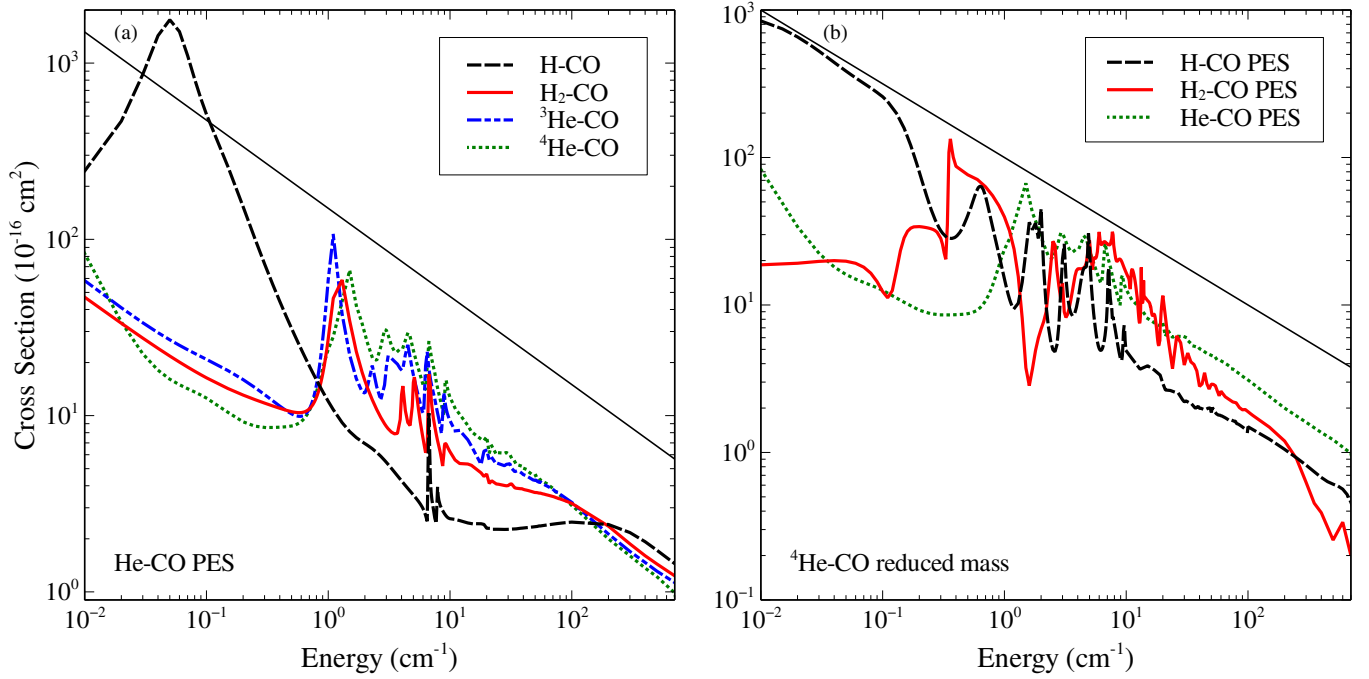
Figure 2. (a) Cross sections of the colliders H, para-H_2, ^{3}He, and ^{4}He on the He–CO PES for the $j = 1 \rightarrow 0$ transition. (b) Cross sections for the same transition using the ^{4}He–CO reduced mass on the H–, H_2–, and He–CO PESs. The straight solid line indicates a $1/v$ cross section dependence.
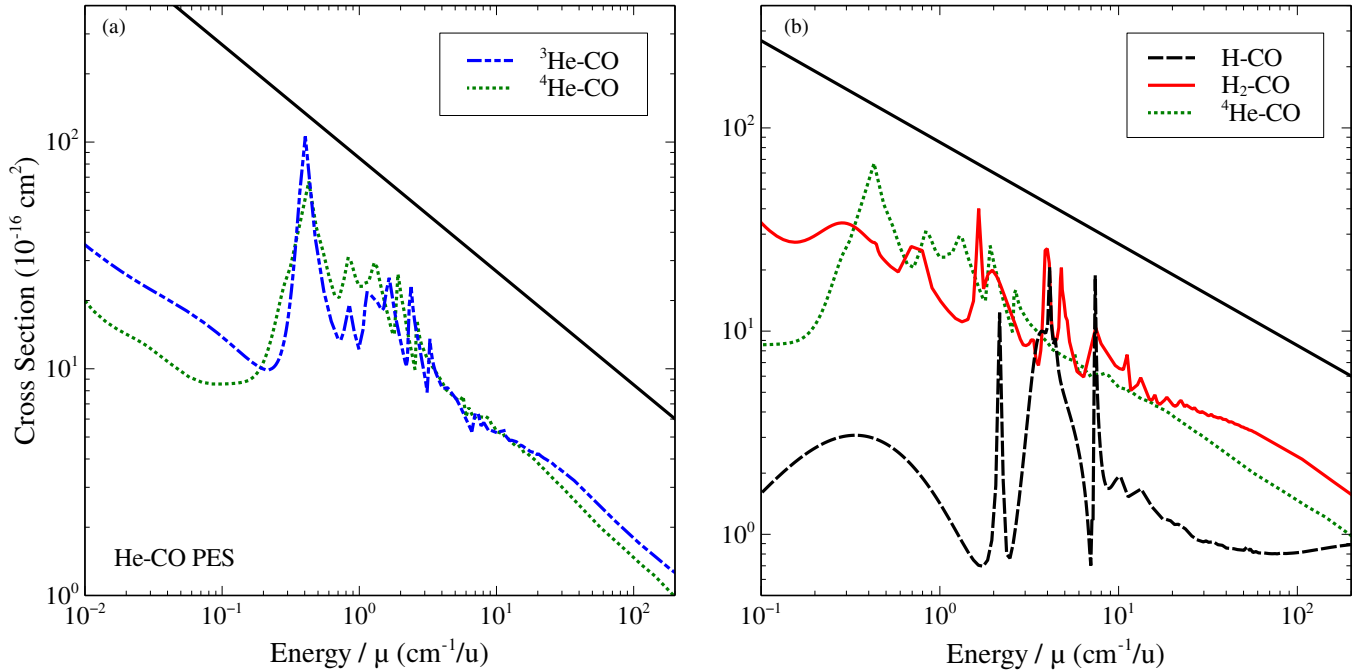(A color version of this figure is available in the online journal.)

Figure 3. Cross sections for the $j = 1 \rightarrow 0$ transition as a function of kinetic energy/μ for (a) the colliders ^{3}He and ^{4}He on the He–CO PESs. (b) Calculated cross sections on their respective PESs. The straight solid line indicates a $1/v$ cross section dependence.
(A color version of this figure is available in the online journal.)

will be relatively independent of both T and μ (i.e., $a = -1$, $b = 0$). The H cross section is smaller due to a considerably different PES structure (see Shepler et al. 2007).

As possible intermediate methods between scaling and explicit calculations, we attempted four other approaches to obtain predictions for para-H_2 as depicted in Figure 4. First, considering the differing PESs, a possible scaling is obtained from the ratio of the reduced potentials $\mu_X \varepsilon_X$ (Joachain 1979), where

ε_X is the van der Waals well depth of the PES. However, the H_2 rate coefficients for the $1 \rightarrow 0$ transition are overestimated (see below). Second, explicit scattering calculations using μ_{H_2} on the He–CO PES gave rates in reasonable agreement with the explicitly calculated H_2–CO results. Third, multiplying these rates by the ratio of the well depths alone (a factor of $\sim$3.9, not shown), and by the ratio of reduced potentials again overestimated the H_2 rate coefficients. Finally, cross sections using
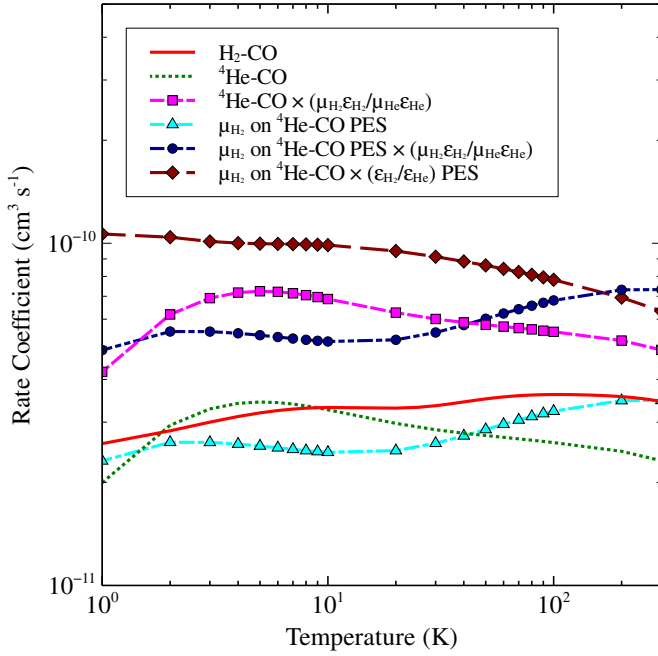
Figure 4. Rate coefficients for the de-excitation of CO($j = 1$) with the colliders He and para-H$_2$ and estimated values for H$_2$ via possible scaling relations; see the text for discussion. $\varepsilon_{H_2} = 93.1$ cm^{-1} (Jankowski & Szalewicz 2005) and $\varepsilon_{He} = 23.7$ cm^{-1} (Heijmen et al. 1997).

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μ_{H_2} on the He–CO PES scaled to match ε_{H_2} were computed, but the resulting rate coefficients overestimated the explicit H$_2$–CO rates. Of these, the second approach, which used the simpler two-dimensional He–CO PES with μ_{H_2} appears to give the best results, but still requires new scattering calculations.

Considering the above findings, we arrive at the two most promising scaling options. In the first case, Equation (6) can

be applied above $\sim$10–50 K when $k_{j \to j'}(T)$ is known for He. The lower limit can be estimated with knowledge of ε_X which is roughly equal to the upper kinetic energy limit of the quasibound resonances (see Figures 2 and 3). For example, if $b = 0$ ($a = -1$), the rate coefficients are independent of T and μ_X, as opposed to Equation (2). Of course, B is assumed to be the same for He and H$_2$ colliders.

The second option is appropriate for $T \lesssim$ 10–100 K where rate coefficients are highly sensitive to quasibound resonances. These resonances may partially be accounted for by scaling via the ratio of the reduced potentials $\mu_X \varepsilon_X$, as discussed above, with a phenomenological exponent C according to

$$k_{j \to j'}^{Z}(T) = \left(\frac{\mu_Z \varepsilon_Z}{\mu_Y \varepsilon_Y} \right)^{C} k_{j \to j'}^{Y}(T). \qquad (7)$$

Numerical values for the van der Waals well depths of the interaction PESs are generally available from experimental and theoretical work in the chemical physics community (see Radzig & Smirnov 1980). This scaling option was explicitly tested for 15 Δj transitions using He–CO (Cecchi-Pestellini et al. 2002) and H$_2$–CO (Yang et al. 2010) theoretical data. The exponent C was optimized to minimize the scaling residuals from $5 \sim 500$ K. Figures 5(a) and (b) shows that both the $2 \to 0$ and $5 \to 4$ rate coefficients scaled via the reduced-potential method give the best estimates. In fact, even Δj transitions scaled by the reduced-potential ratio with exponent $C \sim 0.7$–1.3 give good predictions for H$_2$–CO rate coefficients.

Although standard reduced-mass scaling reproduces the H$_2$–CO data for odd Δj transitions more accurately than even Δj transitions, the agreement is fortuitous. Reduced-potential scaling, on the other hand, with $C \sim 0.0$–0.4 for odd Δj transitions, shows improvement of the predictions *and* is based on the physical properties of the interacting system. We note that near-homonuclear molecules, such as CO, follow propensity rules whereby odd Δj transitions are suppressed compared to even
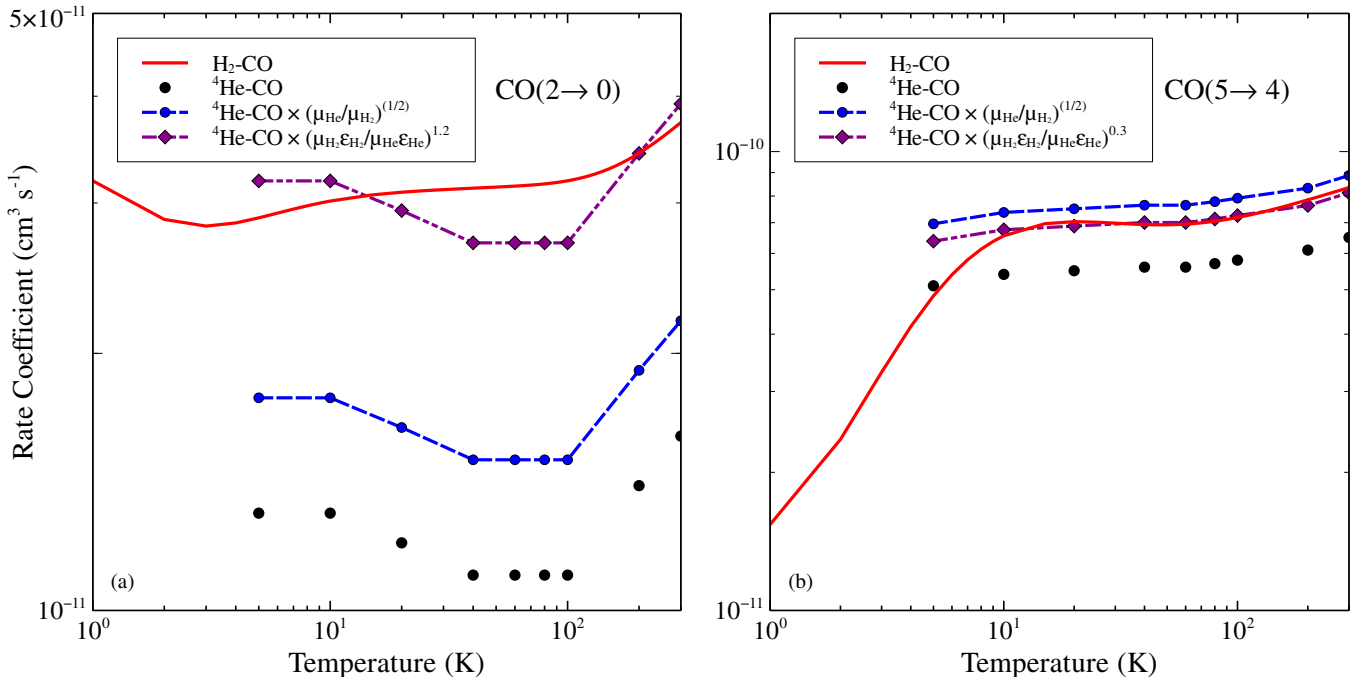

Figure 5. Rate coefficients for the (a) $j = 2 \to 0$ and (b) $j = 5 \to 4$ transitions of CO with H$_2$ (Yang et al. 2010) and He (Cecchi-Pestellini et al. 2002) compared to the predictions of standard reduced-mass scaling and reduced-potential scaling with $C = 1.2$ and 0.3, respectively.

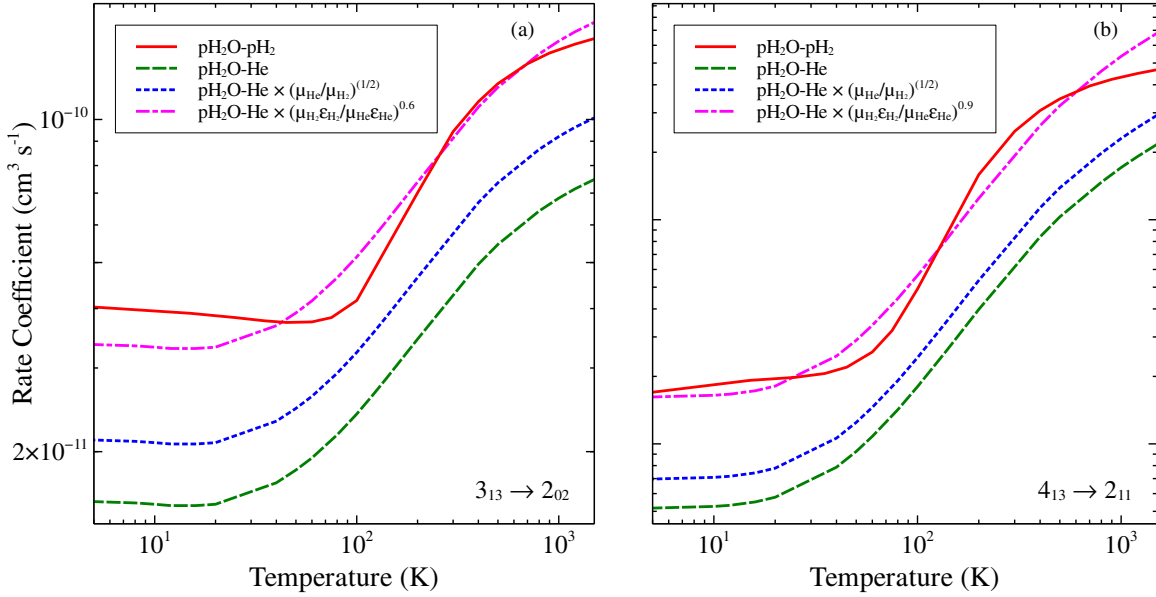(A color version of this figure is available in the online journal.)

Figure 6. Rate coefficients for the de-excitation of $H_2O(j_{k_a k_c})$, (a) $3_{31} \rightarrow 2_{20}$, (b) $4_{13} \rightarrow 2_{11}$, with para-$H_2$ (Dubernet et al. 2009) and He (Yang et al. 2013a) compared to standard reduced-mass scaling and reduced-potential scaling with $C = 0.6$ and 0.9, respectively. The water well depths are $\varepsilon_{He} = 34.4$ cm^{-1} (Patkowski et al. 2002) and $\varepsilon_{H_2} = 221.9$ cm^{-1} (Faure et al. 2005).

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Δj transitions, and it seems the dichotomy of the phenomenological exponent C expresses this propensity.

To determine the accuracy of the new reduced-potential scaling approach, we calculated the normalized root-mean-square deviation (NRMSD), σ_{norm}, of the H_2 rate coefficient predictions for both standard reduced-mass scaling and reduced-potential scaling, given by

$$\sigma_{norm} = \frac{\sqrt{\frac{\sum_{T=i}^{N} (k_{scale}(T) - k_{calc}(T))^2}{N}}}{k_{max} - k_{min}}, \qquad (8)$$

where N is the number of temperature data points and k_{max} and k_{min} are the values of the maximum and minimum rate coefficients, respectively. The resulting percentage indicates the residual variance between the calculated H_2 rate coefficients, k_{calc}, and those scaled from He, k_{scale}. Table 1 lists these values for 15 transitions of CO. There is a remarkable improvement in reduced-potential scaling predictions for even Δj transitions. Odd Δj transitions also show improved predictions of reduced-potential scaling over standard reduced-mass scaling, albeit less so. These odd Δj transitions exhibit the broadest range in rate coefficients and can vary more than an order of magnitude across the temperature range 2–500 K, whereas rate coefficients for the even Δj transitions are primarily flat across this range. Hence the odd Δj transitions contain a larger residual variance.

Figures 6(a) and (b) gives an example of a similar study of reduced-potential and standard reduced-mass scaling for H_2O to due He and para-H_2 collisions. From a survey of 32 transitions, the dominant transitions which obey the propensity rules $|\Delta j| = |\Delta k_a| = |\Delta k_c| = 1$ are reasonably reproduced by the reduced-potential approach with $C \sim 0.6$–0.8, while the subdominant transitions $|\Delta j| = 1$, $\Delta k_a = 0$, $\Delta k_c = \pm 2$ or $\Delta k_a = \pm 2$, $\Delta k_c = 0$ and $|\Delta j| = 2$, $\Delta k_a = 0$, $\Delta k_c = \pm 2$ or $\Delta k_a = \pm 2$, $\Delta k_c = 0$ extend this range to $C \sim 0.5$–1.2. Cases which are reproduced best with $C \lesssim 0.5$ or $\gtrsim 1.2$ typically correspond to weak transitions with rate coefficients 2–3 orders

Table 1
The Optimized Values of C and Their Respective Normalized Root-mean-square Deviations (NRMSDs) for Collisional De-excitation Transitions of CO with H_2 and He Scaled Via the Standard Reduced-mass (rm) and new Reduced-potential (rp) Methods

Δj	$j \rightarrow j'$	C	NRMSD$_{rm}$	NRMSD$_{rp}$
Even	2–0	1.2	138.60	17.95
$\cdots$	3–1	1.2	125.31	18.04
$\cdots$	4–2	1.2	133.00	17.64
$\cdots$	5–3	1.3	141.07	25.73
$\cdots$	4–0	0.9	50.16	9.06
$\cdots$	5–1	0.7	28.57	4.07
Odd	4–3	0.4	27.41	26.63
$\cdots$	3–2	0.4	34.38	34.99
$\cdots$	2–1	0.4	40.07	40.89
$\cdots$	5–4	0.3	31.62	18.81
$\cdots$	1–0	0.3	52.43	41.02
$\cdots$	5–0	0.1	20.06	13.79
$\cdots$	4–1	0.1	35.88	22.23
$\cdots$	3–0	-0.2	38.91	29.48
$\cdots$	5–2	-0.2	38.59	34.45

of magnitude smaller than the dominant transitions so that errors in their prediction are of less significance.

The reliability of the reduced-potential scaling method was again addressed by computing the NRMSD and comparing it to the NRMSD of standard reduced-mass scaling. Figure 7 compares the NRMSD for both methods for each transition of H_2O. Predictions from reduced-potential scaling exhibit less residual variance in all 32 transitions, with a mean of 35% or less, and clearly demonstrate the superiority of the new reduced-potential scaling method.

Finally, Figure 8 shows the two scaling methods for H–CO collisions, where the reduced potential with $C = 0.9$ rather than standard reduced-mass scaling more accurately predicts the calculated rate coefficients. While additional studies of reduced-potential scaling on a variety of other molecules are
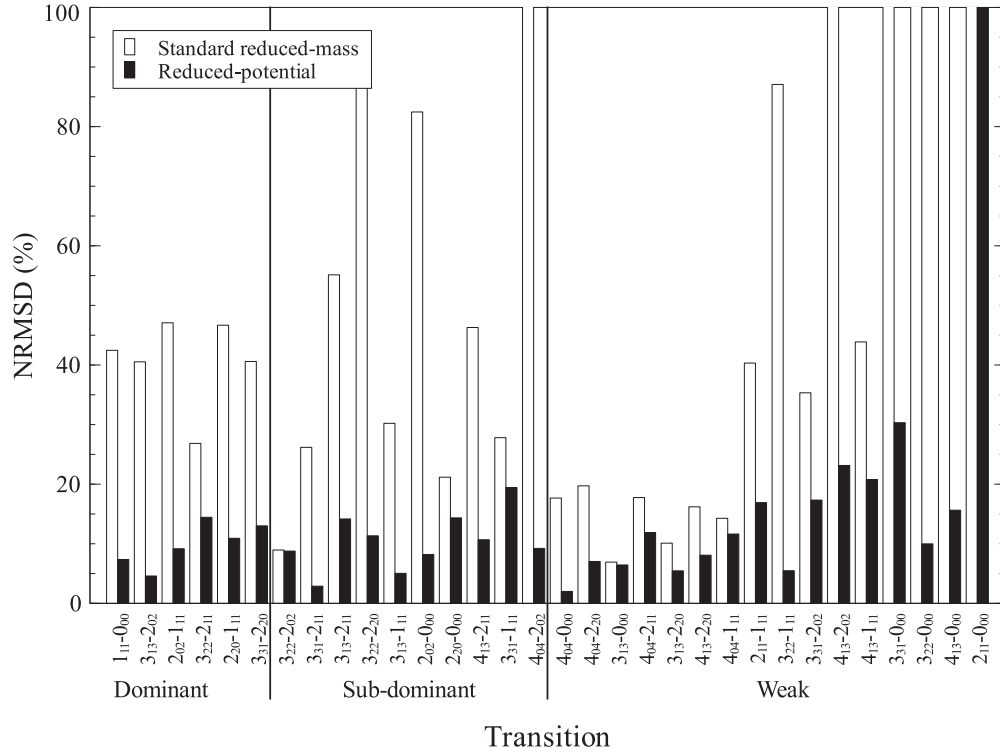
Figure 7. Normalized root-mean-square deviation (NRMSD) in standard reduced-mass scaling and reduced-potential scaling for 32 transitions of H_2O, truncated at 100%. The dominant, sub-dominant, and weak transitions are further organized from left to right in increasing values of the exponent C.
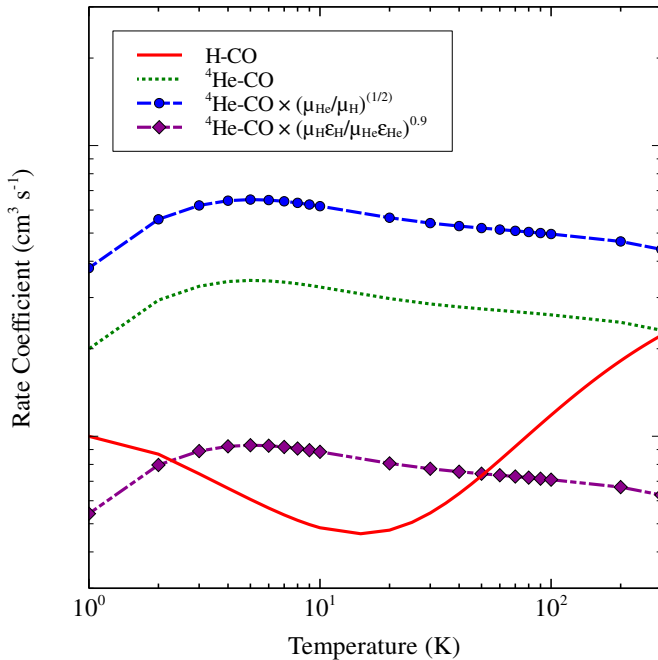
4. CONCLUSION

Rotational inelastic transitions for collisions of H, para-H_2, ^{3}He, and ^{4}He with CO ($j = 1$) using three PESs were computed to study the cross section dependence on reduced mass μ and interaction potential with the goal of gaining insight into rate coefficient scaling. Although earlier investigations indicated that scaling via the ratio of the square root of reduced masses gave reasonable estimates for collisional rate coefficients, the current study, shows that this agreement was fortuitous (see also Schaefer 1990 for similar findings for HD). The constant factor of ~1.4 frequently used to predict the rate coefficients of para-H_2 from that of He generally lead to inaccurate results due to the fact that the underlying assumptions are not valid. Scaling by this standard reduced-mass relation is therefore not recommended. Two alternative scaling approaches are proposed. In the first case, if the inelastic cross section can be represented by an analytical function of the relative velocity, then an exact rate coefficient scaling exists as a function of T and μ, valid for all collision systems. A second approach, which accounts for the contribution of low-energy quasibound resonances, is based on ratios of the product of μ with the PES well depth. Preliminary testing of the reduced-potential method in conjunction with known propensity rules for CO and H_2O gives reasonable predictions. While these two approaches may lead to mathematically and physically reasonable scalings, it is only through explicit calculation and/or measurements that reliable inelastic rate coefficients can be obtained. The improved scaling approaches proposed here may provide useful estimates until such explicit data become available.

Figure 8. Rate coefficients for the de-excitation of CO($j = 1$) with H (Yang et al. 2013b) and He compared to standard reduced-mass scaling and reduced-potential scaling with $C = 0.9$.

(A color version of this figure is available in the online journal.)

needed and in progress, the cases studied here suggest that the approach can reasonably predict rate coefficients for dominant transitions with $C \sim 0.8$, while $C < 0.4$ can account for weak transitions with the partitioning predicted from known propensity rules.

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