Universal behavior of diatomic halo states and the mass sensitivity of their properties

A Owens\textsuperscript{1,2} and V Špirko\textsuperscript{3,4}

\textsuperscript{1} Center for Free-Electron Laser Science (CFEL), Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, D-22607 Hamburg, Germany
\textsuperscript{2} The Hamburg Center for Ultrafast Imaging, Universität Hamburg, Luruper Chaussee 149, D-22761 Hamburg, Germany
\textsuperscript{3} Institute of Organic Chemistry and Biochemistry, Flemingovo nám. 2, 166 10 Prague 6, Czechia
\textsuperscript{4} Department of Chemical Physics and Optics, Faculty of Mathematics and Physics, Charles University in Prague, Ke Karlovu 3, CZ-12116 Prague 2, Czechia

E-mail: vladimir.spiroko@marge.uochb.cas.cz

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Abstract

The scattering and spectroscopic properties of molecular halo states can serve as sensitive probes of the constancy of the electron-to-proton mass ratio $\beta = m_e/m_p$. Since halo states are formed by resonant $s$-wave interactions, their properties exhibit universal correlations that are fairly independent of the interactions at short distances. For diatomic molecules, these properties depend on a single-parameter only, and so this ‘universality’ means that all the characteristics of a diatomic halo state can be determined with high precision if only one-parameter is accurately known. Furthermore, this knowledge can be used to establish the respective property mass sensitivities for investigating the stability of $\beta$. Here, we show for the halo states of the helium dimers that the relationship between the probed properties and their mass sensitivity can be derived from numerically exact solutions of suitable radial Schrödinger equations for a set of effective potential energy curves. The resulting relations exhibit a weak dependence on the short-range part of the used potentials and a near-negligible dependence on the ‘higher-order’ nonadiabatic, relativistic, quantum electrodynamical and residual retardation effects. The presented approach is thus a robust alternative to other literature approaches, particularly in cases where a lack of experimental data prevents an accurate interaction potential from being determined.

Keywords: interatomic and molecular potentials, quantum halo states, molecular physics

(Some figures may appear in colour only in the online journal)

1. Introduction

The formation of a quantum halo state is a phenomenon associated with loosely bound particles held in short-range potential wells. Halo states extend over an unusually large space and many of their properties hinge on the tail of their wavefunctions exhibiting unusual scattering properties \cite{1}. For instance, they allow for the formation of three body systems exhibiting the Efimov effect \cite{2}; the long-range asymptote of the wavefunction of such a state implies quite distant correlations which are relevant for the formation of Bose–Einstein condensates \cite{3} and sympathetic cooling \cite{4}. The actual scattering is characterized by an $s$-wave scattering length that is much larger than the range of the particle interactions. It is therefore conceivable that the scattering length $a$ and its related properties, such as the binding energy $D_0$ or the average value of the internuclear separation $\langle R \rangle$, can exhibit highly anomalous mass dependencies, thus serving as promising probes of a possible variation of the electron-to-proton mass ratio $\beta = m_e/m_p$. Interestingly, as noted by Abraham \textit{et al} \cite{5} and analyzed in detail by Chin and Flambaum \cite{6}, this dependence can be dramatically enhanced in collisions of atoms near narrow Feshbach resonances.
The development of laser-cooling techniques enables long-range halo states to be studied with very high spectral resolution, for example, using photoassociation spectroscopy [7]. Moreover, as was recently shown for He$_2$ in its ground electronic state [8], the square $|\Psi|^2$ of the wavefunction of the halo state can be measured by recording a large number of Coulomb explosion events using cold target recoil ion momentum spectroscopy [9]. Since the properties of halo states are expected to be extremely sensitive to tiny variations of the interaction potential, we have performed calculations for the halo states of the ground and excited $^5\Sigma^+_g$ electronic states of the helium dimers, for which precise theoretical interaction potentials [10, 11] and appropriate experimental bond lengths and binding energies are available. For the electronic ground state these data were obtained by molecular beam diffraction from a transmission grating [12] and from the above mentioned Coulomb explosion experiment [8]; while for the $^5\Sigma^+_g$ electronic state these data were obtained from a two-photon photoassociation experiment [13].

2. Methods

In our approach, the potentials are used to generate relationships between the scattering properties of the probed states and their actual values, from which the mass sensitivities can be determined. This is done through numerical solution of the radial Schrödinger equation for a set of effective potentials $V^{\text{eff}}(R)$, obtained by scaling the interaction potential $V(R)$ with a constant scaling factor $f$ (for further details see [14]),

$$V^{\text{eff}}(R) = f \cdot V(R).$$

The scaled potentials are then utilized to establish the $f$-dependence of the scattering properties, and by combining these dependencies, a direct relationship between the probed properties can be found. For example, establishing the two relations $D_0 = D_0(f)$ and $a = a(f)$ for a set of values of $f$, e.g. $f_i$ ($i = 1$, 2, …), allows us to obtain the pairs $(D_0(f_i), a(f_i))$, which subsequently defines the one-to-one relations $a = F(D_0)$.

Calculations were performed using the effective vibrational Hamiltonian of Herman and Asgharian [15] for nuclear motion in $^1\Sigma$ state molecules,

$$H_{\text{eff}} = -\frac{\hbar^2}{2m} \frac{d}{dR} \left(1 + \beta g_v(R)\right) \frac{d}{dR} + V_{\text{ad}}(R) + V'(R),$$

where $m$ is the appropriate nuclear reduced mass, $V_{\text{ad}}$ is the ‘adiabatic’ part of the molecular potential energy function (assumed to include Born–Oppenheimer, adiabatic, relativistic, quantum electrodynamic (QED) and residual retardation terms), and the terms $V'(R)$ and $g_v(R)$ account for non-adiabatic effects. The so-called vibrational $g_v$-factor is fixed to its ab initio value, and the effective potential energy function $V = V_{\text{ad}}(R) + V'(R)$ is determined either from first principles or from fitting to the available experimental data.

The mass sensitivity of the probed properties, for example, the binding energy $D_0$ and the scattering length $a$,

are described by the following expressions,

$$K_\beta = \beta \frac{dD_0}{D_0 d\beta},$$

$$T_\beta = \beta \frac{da}{a d\beta}.$$
very accurately determined in experiment. The thermo-
physical properties can therefore be checked, and perhaps
predicted more accurately, using the experimental values of
$D_0$ and $a$ and a key aspect in this process is the interaction
potential.

The best available potential for the helium dimer was
recently computed \cite{11} and possesses submillikelvin
uncertainties. The potential accounts for all relevant ‘higher-
order’ effects (adiabatic, relativistic, QED and retardation
corrections), allowing for a direct and detailed analysis of
each contribution. Calculations with these potentials, repre-
sented by table 2 of \cite{11} and table 5 of \cite{10}, reveal the
following important facts (note that we adopt the same
notation here as the aforementioned tables): (a) the most

Figure 1. Top panels: the potential energy function ($V$) and square of the wavefunction ($|\Psi|^2$) of the highest vibrational states of the helium dimers. The interaction potentials are from \cite{10, 11}. Middle panels: the scattering length ($a$) versus binding energy ($D_0$) for the bound state of $^4\text{He}_2$ in the ground electronic state. The curve was obtained by scaling the $V + V_{\text{ret}}$ theoretical potential of \cite{11}. $a_{\text{Vret}}$ and $a_{\text{Grisenti}}$ represent the values and error bars derived using the latter theoretical potential and from experiment \cite{12}, respectively. The points were obtained using the remaining theoretical potentials of \cite{11} and the empirical ‘HFDHe2’ and ‘Hurly’ potentials of \cite{25, 26}. The calculations were performed using the atomic (suffix $a$) and nuclear (suffix $n$) masses. Bottom panels: $a$ versus $D_0$ for the highest bound state of $^5\text{He}_2$ in the $^3\Sigma_g^+$ electronic state. The curves were obtained by scaling the $V + \delta V_{\text{ad}} + \delta V_{\text{el}}$ theoretical potential of \cite{10}. The points were obtained using the remaining theoretical potentials of \cite{10, 27} (denoted GLD).
Coulomb explosion measurements, within the error bars of the experimental value deduced from J. Phys. B: At. Mol. Opt. Phys. relativistic effects contribute significantly meaning that the effect of retardation is small, (c) accurately accounting for nonadiabatic effects is possible through adiabatic calculations with atomic mass values (instead of nuclear masses).

Table 1. Scattering lengths $a$ of $^6$He$_2$ in its ground ($^3\Sigma^+_g$) and excited ($^5\Sigma^+_g$) electronic states (in Å).

| Potential | Calc-1 | Calc-2 | Calc-3 | Calc-4 |
|-----------|--------|--------|--------|--------|
| $V_{BO}$  | 87.847 | 88.634 | 86.718 (+3.889, −3.409) | 86.730 (+3.889, −3.410) |
| $V_{BO} + V_{ad}$ | 85.550 | 86.295 | 86.715 (+3.889, −3.409) | 86.730 (+3.889, −3.410) |
| $V_{BO} + V_{ref}$ | 93.827 | 94.730 | 86.715 (+3.889, −3.409) | 86.726 (+3.889, −3.410) |
| $V_{BO} + V_{ad} + V_{ref}$ | 91.194 | 92.045 | 86.715 (+3.889, −3.409) | 86.726 (+3.889, −3.410) |
| $V$ | 90.376 | 91.211 | 86.716 (+3.889, −3.409) | 86.728 (+3.889, −3.409) |
| $V + V_{ref}$ | 90.502 | 91.339 | 86.716 (+3.889, −3.409) | 86.728 (+3.889, −3.409) |
| Hurly | 88.323 | 89.119 | 86.715 (+3.889, −3.409) | 86.727 (+3.889, −3.410) |
| HFDHe2 | 124.304 | 125.909 | 86.723 (+3.889, −3.409) | 86.735 (+3.889, −3.410) |

Note. If not stated otherwise the interaction potentials and notation are from [10, 11], $D_0 = 36.73$ MHz [8] and 91.35 MHz [13] for the ground and excited state of $^6$He$_2$, respectively. Calc-1 and Calc-2: derived from the ‘zero collision energy’ wavefunctions using atomic ($m_{amu} = 4.002603$ 254 13 amu) and nuclear ($m_{amu} = 4.001506$ 179 125 amu) masses, respectively; Calc-3 and Calc-4: evaluated using the ‘$a$ versus $D_0$’ relations with atomic and nuclear masses, respectively. Hurly: obtained using the potential of [25], HFDHe2: obtained using the potential of [26]. GLD: obtained using the potential of [27].

Table 2. Average internuclear separations ($R$) of $^6$He$_2$ in its ground ($^3\Sigma^+_g$) and excited ($^5\Sigma^+_g$) electronic states (in Å).

| Potential | Calc-1 | Calc-2 | Calc-3 | Calc-4 |
|-----------|--------|--------|--------|--------|
| $V_{BO}$  | 45.802 | 46.195 | 45.237 (+1.946, −1.706) | 45.243 (+1.946, −1.706) |
| $V_{BO} + V_{ad}$ | 44.653 | 45.025 | 45.237 (+1.946, −1.706) | 45.243 (+1.946, −1.706) |
| $V_{BO} + V_{ref}$ | 48.793 | 49.244 | 45.234 (+1.963, −1.706) | 45.240 (+1.946, −1.706) |
| $V_{BO} + V_{ad} + V_{ref}$ | 47.475 | 47.901 | 45.234 (+1.946, −1.706) | 45.239 (+1.946, −1.706) |
| $V$ | 47.067 | 47.485 | 45.236 (+1.946, −1.706) | 45.243 (+1.946, −1.706) |
| $V + V_{ref}$ | 47.129 | 47.548 | 45.235 (+1.946, −1.706) | 45.241 (+1.946, −1.706) |
| Hurly | 46.040 | 46.438 | 45.235 (+1.946, −1.706) | 45.241 (+1.946, −1.706) |
| HFDHe2 | 64.026 | 64.827 | 45.242 (+1.946, −1.706) | 45.247 (+1.946, −1.706) |
| $V_{U}$ | 49.289 | 50.012 | 48.366 (10) | 48.370 (9) |
| $V$ | 48.542 | 49.234 | 48.365 (9) | 48.369 (9) |
| $V_{L}$ | 47.827 | 48.489 | 48.363 (9) | 48.367 (9) |
| $V + V_{ad}$ | 47.397 | 48.041 | 48.367 (9) | 48.371 (9) |
| $V + V_{ref}$ | 49.501 | 50.233 | 48.365 (9) | 48.369 (9) |
| $V + V_{ad} + V_{ref}$ | 48.290 | 48.970 | 48.367 (9) | 48.371 (9) |
| GLD | 93.189 | 97.265 | 48.410 (10) | 48.405 (9) |

Note. If not stated otherwise the interaction potentials and notation are from [10, 11]. Calc-1 and Calc-2: derived from the ‘zero collision energy’ wavefunctions using atomic and nuclear masses, respectively; Calc-3 and Calc-4: evaluated using the ‘$a$ versus $D_0$’ relations with atomic and nuclear masses, respectively. Hurly: obtained using the potential of [25], HFDHe2: obtained using the potential of [26]. GLD: obtained using the potential of [27].

rigorous theoretical potential predicts the binding energy within the error bars of the experimental value deduced from Coulomb explosion measurements, (b) the adiabatic and relativistic effects contribute significantly meaning that the effect of retardation is small, (c) accurately accounting for nonadiabatic effects is possible through adiabatic calculations with atomic mass values (instead of nuclear masses). Interestingly, see the middle and bottom panels of figure 1, tables 1 and 2, upon scaling of the interaction potential defined by equation (1), the different potentials all provide closely coinciding relations between the calculated properties. Furthermore, as expected from our previous calculations [14], the same relations are also established for the empirical potentials obtained from fitting to the
thermochemical data [25, 26]. This demonstrates the ‘universality’ in two body systems [30], where the knowledge of only one of the probed scattering properties, in conjunction with a moderately accurate interaction potential, allows for a quantitative prediction of all the remaining properties.

The ‘universality’ of the simple scaling defined by equation (1) is also deeply reflected in the fitting of the experimental Coulomb explosion data for the helium dimer wavefunction $\Psi$ of [8]. In figure 2, the top panels illustrate the dispersion of the theoretical $\Psi$ values evaluated using different interaction potentials; the middle panels show the agreement of the theoretical $\Psi$ values obtained with potentials scaled to provide the experimental binding energy $D_0 = 36.73$ MHz, deduced from experimental data corrected for electron-recoil effects; the bottom panels reproduce the uncorrected experimental data using potentials scaled to provide the best least squares fit of the data; $f = 1.004 227 56, 1.003 394 92, 1.006 186 42, 1.005 349 27, 1.005 125 56, and 1.004 176 15 for $V_{BO}$, $V_{BO} + V_{rel}$, $V_{BO} + V_{rel} + V_{ret}$, $V_{BO} + V_{rel} + V_{ret}$, $V_{BO} + V_{rel} + V_{ret}$, respectively. All calculations were performed using atomic masses. The right hand side panels represent details of the fits.
Figure 3. The squares of the $^3$He$_2$ wavefunctions ($|\Psi|^2$) in the $^5\Sigma_g^+$ electronic state evaluated using the original potentials of [10, 27] (top panels) and their versions scaled so that they provide the experimental binding energy of [13] (bottom panels).

Figure 4. Left panel: the scaled binding energy $\epsilon_s = 2mD_o\alpha_0^2/\hbar^2$ as a function of $1/a_{\text{th}} = \beta_0/a$ ($\beta_0 = (2mC_0/\hbar^2)^{1/4}$). Analytic - results obtained using the analytic formula of [20] (see equation (14) of [20]). Scaled — numerically exact calculations. Effective range — results obtained using the $V + \delta V_{ad} + \delta V_{el}$ potential energy of [10]. Right panel: comparison of the $'a$ versus $D_o'$ relations obtained for the $^5\Sigma_g^+$ state. $V_{\text{scaled}}$ - results obtained using the $V + \delta V_{ad} + \delta V_{el}$ theoretical potential of [10]. $V_{\text{mod}}$ - results obtained using the $V + \delta V_{ad} + \delta V_{el}$ potential with discarding the $C_11$ and $C_{12}$ contributions. $V_{\text{HST}}$ - results obtained using an effective interaction potential consisting of a hard-sphere with the $-C_6/R^6$ attractive tail (see equation (4) of [21]) and atomic masses.
46 MHz, giving insight into the role of electron-recoil effects).

Generally speaking, the close agreement of the scaled $\Psi$ values is also present in other halo states, for example, the spin-polarized helium atoms in the $2S$ metastable state (see figure 3). Importantly, as seen in the previous figures, the fitted wavefunctions closely coincide over the whole interval of internuclear separations, demonstrating the robustness of the scaling approach defined by equation (1) over the whole range of relevant binding energies and scattering lengths.

In figure 4, we see that for small binding energies the presented scheme is in excellent agreement with the results obtained using quantum defect theory, which is usually employed in the literature [20, 21]. However, for higher binding energies associated with scattering lengths tending to $-\infty$, these approaches do not provide reliable asymptotes and thus fail to provide reliable mass sensitivities in these energy regions, particularly for scattering lengths exhibiting a non-linear and discontinuous energy dependence. These regions appear to be particularly promising for probing the mass sensitivity of the scattering properties (see figure 5), however, as shown in table 3, the dimensionless scaling of the ‘global’ interaction potential should be adequate for the entire energy region. It should be stressed that only globally accurate potentials appear to be adequate for this critical region.

4. Conclusions

The scattering properties of diatomic halo states can be extracted from one-parameter only $f$-relationships. These relations were derived from numerically exact solutions of the radial Schrödinger equation for a set of effective potentials, obtained by a multiplicative scaling of the ‘generic’ interaction potential. The calculated scattering properties are as accurate as their unscaled $f = 1$ counterparts, which are derived from the best available interaction potentials. Furthermore, the predicted values appear somewhat independent of the underlying potential, suggesting that any moderately accurate potential can be utilized in the presented approach. As anticipated, the mass sensitivities of the scattering lengths and of the related properties grow significantly when increasing the classically forbidden part of the halo state wavefunction. For example, the sensitivities of the delocalized state of $^4\text{He}^3\text{He}$ are about one order of magnitude larger than those relating to a less delocalized state of $^3\text{He}_2$. 
Table 3. The mass sensitivity coefficients $K_n$ and $T_n$ of $^4$He$_2$ in its ground ($\Sigma^+_g$) and excited ($\Sigma^+_e$) electronic states.

| Potential       | $K_1^g$ | $K_1^e$ | $K_2^g$ | $K_2^e$ | $T_1^g$ | $T_1^e$ | $T_2^g$ | $T_2^e$ |
|-----------------|---------|---------|---------|---------|---------|---------|---------|---------|
| $\Sigma^+_g$    | $V_{\text{coll}}$ | 66.611  | 67.228  | 65.726  | 65.735  | −32.35  | −32.66  | −31.91  | −31.91  |
|                 | $V_{\text{coll}} + V_{\text{ad}}$ | 64.809  | 65.393  | 65.725  | 65.734  | −31.45  | −31.75  | −31.92  | −31.92  |
|                 | $V_{\text{coll}} + V_{\text{el}}$ | 71.374  | 72.083  | 65.787  | 65.796  | −34.73  | −35.09  | −31.94  | −31.95  |
|                 | $V_{\text{coll}} + V_{\text{ad}} + V_{\text{el}}$ | 69.305  | 69.974  | 65.787  | 65.796  | −33.70  | −34.03  | −31.94  | −31.95  |
|                 | $V$     | 68.637  | 69.292  | 65.763  | 65.773  | −33.37  | −33.69  | −31.93  | −31.94  |
|                 | $V + V_{\text{el}}$ | 68.744  | 69.402  | 65.771  | 65.780  | −33.42  | −33.75  | −31.93  | −31.94  |
| $HST$           | 67.035  | 67.660  | 65.772  | 65.781  | −32.57  | −32.88  | −31.94  | −31.94  |
| $HFDHe$         | 94.936  | 96.193  | 65.532  | 65.541  | −46.50  | −47.13  | −31.82  | −31.82  |

Note. If not stated otherwise the interaction potentials and notation are from $[10, 11]$. $D_0 = 36.73$ MHz $[8]$ and 91.35 MHz $[13]$ for the ground and excited state of $^4$He$_2$, respectively. $K_1^g/T_1^g$ and $K_2^g/T_2^g$ derived from the ‘zero collision energy’ wavefunctions using atomic and nuclear masses, respectively; $K_1^e/T_1^e$ and $K_2^e/T_2^e$ evaluated using ‘$a$ versus $D_0$’ with atomic and nuclear masses, respectively. $HST$: obtained using the potential of $[25]$, $HFDHe$: obtained using the potential of $[26]$, $GLD$: obtained using the potential of $[27]$. $HST$: calculations performed using the $V_{\text{EFF}}$ effective potential of $[21]$ with $r_0 = 3.8$ Å and $C_6 = 276.68$ a.u. $[10]$. In the case of $^4$He$_2$ and $^3$He$^3$He, where $D_0$ is not available, $V_{\text{EFF}}$ was scaled so that it provides the same $T_2^g$ as its $ab$ initio counterpart $V$.

Most importantly, a single-parameter scaling based on equation (1) allows for the close fit of the square of the experimental halo state wavefunction $|\psi|_0^2$, thus reflecting the universality of quantum diatomic halo states of diatomic systems (see, e.g. $[30]$). This universality means that all the parameters characterizing the low-energy scattering of atoms can be determined with high precision if only one of these characteristics is accurately known. Moreover, scattering properties and their mass sensitivities, which appear promising for investigating the stability of the electron-to-proton mass ratio $\beta$, can be fully determined. All one requires is a potential energy curve of moderate accuracy and an accurate experimental value for a pertinent scattering property. In principle, unlike the semiclassical $[19]$ or quantum defect theories $[32, 33]$ which can be viewed as the standard alternatives for deriving the ‘$a$ versus $D_0$’ relations, the scaling described by equation (1) can be used straightforwardly for any type of long-range potential asymptote.

Currently, the best (spectroscopic) measurements of the scattering length can probe a variation of $\beta$ at the level of $10^{-13} \ldots 10^{-16}$ yr$^{-1}$ $[6]$. Given that the scattering phase shift $\delta_0$ can be measured with a precision that yields scattering lengths with 1ppm accuracy $[34]$ (see also $[35]$), one can expect to investigate temporal variations of $\beta$ at the level of $10^{-15} \ldots 10^{-18}$ yr$^{-1}$. The presented approach can cope with this accuracy by providing accurate mass sensitivities even in cases where a lack of spectral data prevents the use of standard approaches. However, it should be noted that the accuracy of the experimental setup will dictate future investigations of $\beta$, and large sensitivity coefficients may not always be transferable to experiment.
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ORCID iDs

A Owens https://orcid.org/0000-0002-5167-983X

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