



Quantum dynamics calculations for $e^+ + \text{LiH} \rightarrow \text{Li}^+ + [\text{H}^-; e^+]$ dissociative positron attachment using a pseudopotential model

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ABSTRACT

We theoretically discuss the dynamics of the chemical reaction process induced by positron attachment, $e^+ + \text{LiH} \rightarrow \text{Li}^+ + [\text{H}^-; e^+]$. Simple model pseudopotentials were developed on the basis of the electrostatic interaction between e^+ and LiH with a large dipole moment, where some numerical parameters were chosen to qualitatively reproduce the previous-reported *ab initio* positron-bound adiabatic potential energy curve of $[\text{LiH}; e^+]$. Using the developed pseudopotential models, we have performed time-independent quantum scattering calculations using the hyperspherical coordinate. Although the main scattering processes of the $e^+ + \text{LiH}$ collision are rotational and vibrational excitation processes, it is found that the calculated probabilities for the dissociative positron attachment (DPA) process are not very small within the present pseudopotential framework. This suggests that the DPA process to LiH can be observed in the future experiments.

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1. Introduction

Positrons have been widely used to characterize negatively charged defects or vacancy defects in materials [1,2] and to probe tumors through positron emission tomography [3,4]. However, the detailed microscopic-level mechanisms of the interactions between positrons and molecules are not yet fully understood. This is in high contrast with the fact that the interaction between electrons and molecules has been extensively studied experimentally and theoretically [5]. Recent advances in the development of low-energy positron beam techniques have opened a new avenue for studying chemical dynamics induced by positron collisions at an atomic level. For example, Surko and coworkers have measured positron binding energies (or positron affinities) for a number of molecules, including organic polyatomic compounds, through vibrational Feshbach resonances using well-controlled positron beams [6–9]. In addition to this experimental development, the interaction between positrons and simple molecules has been studied theoretically using electronic structure theory including positron-electron correlation effects [10–29]. For example, Tachikawa and coworkers developed the multi-component molecular orbital method [10–13] and quantum Monte Carlo method

[20,21] to calculate positron binding energies for various molecules. More recently, the any-particle molecular orbital method [30] has been used to calculate positron binding energies in molecules [16,17]. Moreover, many positron molecular scattering calculations with fixed molecular structures have been performed, although these calculations do not directly provide positron binding energies [31,32]. All these theoretical studies can provide important basis for understanding the chemical dynamics processes induced by positron collisions.

In this work, we focus on the dynamics of LiH dissociation induced by positron attachment by using a computational approach. Previous works have already shown that attractive electrostatic interactions formed by the permanent dipole moment of a molecule play an important role in positron binding [33]. Because alkali metal hydride diatomic molecules (LiH, NaH, and KH) generally have large dipole moments due to their ionic bonding character, positron binding has been frequently investigated in these molecules [18–29]. The first study on the $[\text{LiH}; e^+]$ system was performed by Kurtz and Jordan using the Hartree–Fock method [18]. Subsequently, several theoretical studies were performed. Mella et al. reported a potential energy curve for the positron-bound LiH quasimolecule, $[\text{LiH}; e^+]$, in the $R(\text{Li-H})$ internuclear distance range of 2–12 a_0 using the quantum Monte Carlo method [23]. They found that the equilibrium distance of $[\text{LiH}; e^+]$ is longer than that of the neutral LiH molecule by about 0.2 a_0 . This indicates that adding an excess positron to LiH affects the bonding character and

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decreases the bond strength. Gianturco et al. [25] reported a similar potential energy curve calculated with the configuration interaction approach, although the positron affinity at the neutral LiH equilibrium distance was about 20% smaller than that of Mella et al. Oyamada and Tachikawa have recently reported a smooth potential energy curve for $[\text{LiH}; e^+]$ obtained by multi-component full-configuration interaction calculations [29]. Their calculation somewhat underestimated the positron binding energy at the equilibrium LiH distance compared with previous results [18–28].

It should be mentioned that the potential energy curve for the positronic $[\text{LiH}; e^+]$ quasimolecule asymptotically approaches the $\text{Li}^+ + [\text{H}^-; e^+]$ dissociation limit, where $[\text{H}^-; e^+]$ is the atomic bound state in which two electrons and one positron are simultaneously bound by a proton nucleus. Alternatively, $[\text{H}^-; e^+]$ can be written as PsH, where a positronium (Ps, bound state of positron and electron) is formed on the H atom. Because the positron binding energy of H^- is larger than the ionization energy of Li [34,35], the energy level of the $\text{Li}^+ + [\text{H}^-; e^+]$ dissociation limit is below the neutral Li + H dissociation limit (see Fig. 2 below). This suggests that dissociative positron attachment (DPA), $e^+ + \text{LiH}(\nu, j) \rightarrow \text{Li}^+ + [\text{H}^-; e^+]$ (ν and j denote vibrational and rotational quantum numbers of LiH, respectively), could be observed if the dissociation occurs within the positron annihilation lifetimes. An e^+e^- pair may annihilate

Table 1

Parameters used in the Model-I and Model-II pseudopotential functions.

Model-I	Model-II
$A = 1.621 \text{ au}$	$\beta_e' = 0.365 a_0^{-1}$ $\beta_e'' = 0.221 a_0^{-1}$
$\alpha = 0.7 a_0^{-1}$	$\alpha = 0.7 a_0^{-1}$
$r_s = 4.5 a_0$	$r_s = 4.0 a_0$ $D_e' = D_e'' = 5.0 \text{ eV}$ $r_e' = r_e'' = 2.0 a_0$

and emit two or three γ -ray photons with lifetimes of ~ 100 ns timescale [36].

In previous positron-molecule scattering calculations, the positron-molecule has been frequently approximated by means of a local potential consisting of the electrostatic potential and the polarization potential [37–39], since full *ab initio* treatment of correlation effects in positron-molecule interactions including nuclear degrees of freedom is generally a difficult problem. Meanwhile, there have been a number of theoretical studies on electron-molecule scattering using a pseudopotential approach [40–44]. It should be emphasized that, in most of those previous studies, a

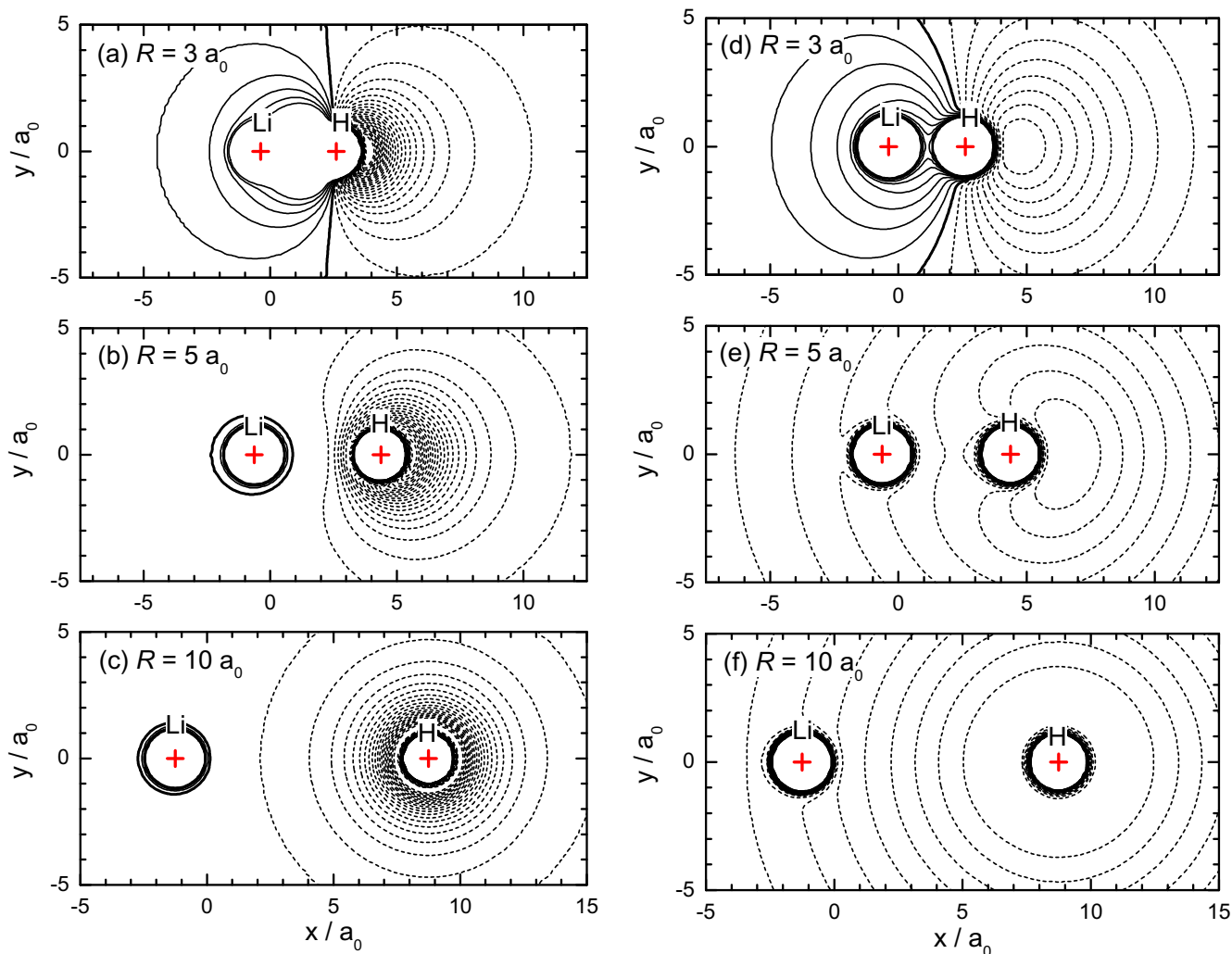


Fig. 1. Contour plots of the interaction energy between e^+ and LiH as a function of the Cartesian positron position at selected values of the LiH distance, where the center of mass of LiH is located at the coordinate origin. The contour increment is set to 1 eV for panels (a)–(c) (Model-I) while 0.5 eV for panels (d)–(f) (Model-II). Positive and negative contours are shown with solid and dashed lines, respectively. Zero energy contours are shown with bold solid lines.

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