

Stability of four-body systems in three and two dimensions: A theoretical and quantum Monte Carlo study of biexciton molecules

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The stability of four-body systems ($m_a^+ m_b^+ m_1^- m_2^-$) in three (3D) and two dimensions (2D) is discussed using accurate numerical results obtained by means of diffusion Monte Carlo calculations. In 3D, we extend our proof of the stability for the class of systems ($m_a^+ m_b^+ m_1^- m_1^-$), showing that they are stable against the dissociation in ($m_a^+ m_1^-$) and ($m_b^+ m_1^-$) for any value of the mass ratio m_a^+/m_b^+ . In 2D, using the ground-state energy of the dipositronium, it is possible to prove that the stability of four-body systems follows the same scenario. We also give upper and lower bounds to the binding energies for the class ($M^+ M^+ m^- m^-$) in 2D, useful to discuss the relative stability of biexciton molecules in semiconductors. [S1050-2947(98)03206-5]

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I. INTRODUCTION

The problem of the stability of few unit-charge systems with respect to dissociation into smaller fragments is of central importance in modern atomic, cluster, particle, and solid-state physics. Although many papers have been published on three- and four-body unit-charge systems in the “real” three-dimensional (3D) world [1–12], few calculations and studies [13–15] have been carried out for the “unphysical 2D Flatland” world, where “Flatland” is the ideal two-dimensional world described by Abbott [16]. However, there are many physical situations where particles are almost effectively moving in a space of lower dimensionality. The most striking examples are the negatively charged exciton [17], the biexciton molecule in semiconductors [13], and systems in high magnetic fields [18]. Due to computational difficulties, more work has been done on three particle systems in three dimensions than on four-particle systems: if an accurate description is needed, i.e., if one is forced to use nonadiabatic explicitly correlated wave functions, passing from three to four particles is not an easy task since matrix elements are quite difficult to compute [13]. This problem becomes even worse when moving from three to two dimensions, due to its mathematical complexity and to the higher correlation energy, explaining the smaller number of papers published on this subject.

In this paper we present numerical results concerning the stability of four-body Coulomb clusters in two and three dimensions. These results enable us to extend our demonstration of the stability of ($m_a^+ m_b^+ m_1^- m_1^-$) in three dimensions [12], and to show that the same stability scenario holds also for the 2D case.

II. 3D CASE

In the following a numerical subscript denotes a negatively charged particle, while an alphabetical subscript de-

notes a positively charged one. The nonrelativistic Hamiltonian operator, in a.u., for the general system ($m_a^+ m_b^+ m_1^- m_2^-$), has the form

$$\mathcal{H} = -\frac{1}{2} \left(\frac{\nabla_1^2}{m_1} + \frac{\nabla_2^2}{m_2} + \frac{\nabla_a^2}{m_a} + \frac{\nabla_b^2}{m_b} \right) + V(\mathbf{R}), \quad (1)$$

where $V(\mathbf{R})$ is the Coulomb interaction potential:

$$V(\mathbf{R}) = \frac{1}{r_{ab}} + \frac{1}{r_{12}} - \frac{1}{r_{1a}} - \frac{1}{r_{1b}} - \frac{1}{r_{2a}} - \frac{1}{r_{2b}} \quad (2)$$

between the four-unit-charge particles, and \mathbf{R} is a point in configuration space.

In a previous paper [12], we showed that the systems ($M^+ m^+ M^- m^-$) remain stable against the dissociation in the two fragments ($M^+ M^-$) and ($m^+ m^-$), as long as

$$0.476 \leq \frac{M}{m} \leq 2.1. \quad (3)$$

Exploiting this result, we were able to show that ($M^+ m^+ m^- m^-$) is stable for any value of the mass ratio M/m , and that the more general class of systems ($m_a^+ m_b^+ m_1^- m_1^-$) is stable for any value of the masses m_a and m_b as long as $m_a \geq m_1$ and $m_b \geq m_1$. Here we show that this result can be extended to include any physical value of the masses $m_a, m_b \geq 1$.

For the general family ($m_a^+ m_b^+ m_1^- m_2^-$), it is possible to define two new quantities [4]

$$\frac{2}{\Delta} = \frac{1}{m_a} + \frac{1}{m_1}, \quad (4)$$

$$\frac{2}{\delta} = \frac{1}{m_b} + \frac{1}{m_2},$$

where, without any loss of generality, we impose $m_a \geq m_b$ and $m_1 \geq m_2$. In his work, Richard [4] proved that if the ratio Δ/δ is within the range of stability of the systems ($M^+ m^+ M^- m^-$), the variational principle implies the stabil-

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ity for the general four-body Coulomb cluster. Simple scaling arguments based on the variational principle allow one to prove that the stability of the systems with $m_1 = 1$ implies the stability of the more general systems having the same mass ratio between positive and negative particles: thus we restrict our demonstration and calculation to the case $m_1 = 1$.

If we pose $m_1 = m_2 = 1$, and define the quantities

$$\begin{aligned}\sigma_a &= \frac{1}{m_a}, \\ \sigma_b &= \frac{1}{m_b},\end{aligned}\quad (5)$$

the ratio Δ/δ can be written as

$$\frac{\Delta}{\delta} = \frac{\sigma_b + 1}{\sigma_a + 1}.\quad (6)$$

Let us define $L > 1$ and $l = 1/L$, respectively, as the upper and lower limits of the range of stability for the systems $(M^+ m^+ M^- m^-)$. From $1/L \leq \Delta/\delta \leq L$, one obtains the system of inequalities

$$\begin{aligned}\sigma_a &\leq L(\sigma_b + 1) - 1, \\ \sigma_a &\geq \frac{\sigma_b - (L - 1)}{L}\end{aligned}\quad (7)$$

that has to be solved to find the range of stability for the system $(m_a^+ m_b^+ m_1^- m_1^-)$. The inequality $\sigma_a \leq (L + 1)\sigma_b - 1$ is satisfied for any $L \geq 1$, since $\sigma_a \leq \sigma_b$. The remaining inequality is satisfied in the range $0 \leq \sigma_b \leq L - 1$, so that if $L \geq 2$ these systems are stable for all the values of $m_a, m_b \geq 1$. This completes the proof since we have already shown [12] that $L \geq 2.1$. The same result was obtained by Varga, Fleck, and Richard [19], and numerical evidence was given. In other words, it can be stated that every time there are two particles of equal mass and charge in a four-body Coulombic system, the stability of the system itself is guaranteed. Well-known examples of this class of clusters are the hydrogen molecule and all its isotopic derivatives, and the four-body muon molecules $pp\mu\mu$, $pd\mu\mu$, $pt\mu\mu$, $dd\mu\mu$, $dt\mu\mu$, and $tt\mu\mu$ [20].

III. 2D CASE

Similar results can also be obtained for the 2D ‘‘Flatland’’ case of four-body Coulomb systems $(m_a^+ m_b^+ m_1^- m_2^-)$, where the same analytical form for the interaction potential between charges is assumed instead of the solution in two dimensions of the Poisson equation. We begin defining the new quantities $\sigma_v = 1/m_v$ for all particles of the cluster, recalling that the ground-state energy for the system $m_a^+ m_1^-$ in two dimensions is $-2/(\sigma_a + \sigma_1)$.

In his work Rebane [21] showed that, in three dimensions,

$$\begin{aligned}E(\sigma_a, \sigma_b, \sigma_1, \sigma_2) &\leq E_{\text{upper}}(\sigma_a, \sigma_b, \sigma_1, \sigma_2) \\ &= \frac{4E^0}{\sigma_a + \sigma_b + \sigma_1 + \sigma_2},\end{aligned}\quad (8)$$

where $E(\sigma_a, \sigma_b, \sigma_1, \sigma_2)$ is the exact ground-state energy of the general system, while E^0 is the ground-state energy of

the reference system with $\sigma_a = \sigma_b = \sigma_1 = \sigma_2 = 1$. This result holds also in two dimensions, since it is obtained using only the scaling properties of the Coulomb potential and of the Laplacian operator. The same is also true for the results obtained by Richard [4], especially for the stability of the general system $(m_a^+ m_b^+ m_1^- m_2^-)$ as a function of the two quantities Δ and δ defined in Eq. (4).

Using Eq. (8), it is possible to define a minimal range of stability for the symmetric systems $(M^+ m^+ M^- m^-)$, simply imposing that the upper bound $E_{\text{upper}}(\sigma_a, \sigma_b, \sigma_1, \sigma_2)$ lies below the energy of the two fragments $(M^+ M^-)$ and $(m^+ m^-)$, i.e., $E_{\text{thr}}^{2D} = -M - m = -1/(\sigma_M) - 1/(\sigma_m)$. One obtains the inequality

$$\left(\frac{\sigma_M}{\sigma_m}\right)^2 + 2(1 + E^0)\frac{\sigma_M}{\sigma_m} + 1 \leq 0,\quad (9)$$

dependent on the value of the ground-state energy E^0 of the reference system. To compute this value, variational and diffusion Monte Carlo (DMC) simulations [23] on the dipositronium molecule in two dimensions were performed. The DMC total energy, extrapolated to a zero time step, is $E^0 = -2.1928(1)$ hartree. This energy is in optimal agreement with the value of -2.192858 hartree computed by Varga using explicitly correlated Gaussians [22]. Using this result and Eq. (9), one obtains

$$0.543 \leq \frac{M}{m} \leq 1.843\quad (10)$$

as the minimal range of stability for the $(M^+ m^+ M^- m^-)$ systems. To prove that the stability scenario present in three dimensions holds also in two dimensions, one has to show that in two dimensions the upper limit is at least equal to 2, and this is easily done using the DMC approach. In fact, for the system $(M^+ m^+ M^- m^-)$ with $M = 2$ and $m = 1$, we obtained a ground-state energy of $-3.1379(14)$ hartree, i.e., a binding energy of $0.1379(14)$ hartree. This is about 100 times the binding energy of the same system in three dimensions, i.e., $0.00140(4)$ hartree [12]. The stability of $(M^+ m^+ m^- m^-)$ in two dimensions follows as a consequence of the stability of $(M^+ m^+ M^- m^-)$ for $M = 2$ and $m = 1$ [12].

The DMC energy of dipositronium in two dimensions can also be exploited to define lower and upper bounds to the binding energy for the class $(M^+ M^+ m^- m^-)$ in two dimensions, a model of biexciton molecules in semiconductors. Using Eq. (8) with $\sigma_a = \sigma_b = \sigma_M$ and $\sigma_1 = \sigma_2 = \sigma_m$, together with the dissociation energy in the two fragments $(M^+ m^-)$ $E_{\text{thr}} = -4Mm/(M + m) = -4/(\sigma_M + \sigma_m)$, for the biexciton binding energy E_{XX} one obtains

$$E_{XX}(M^+ M^+ m^- m^-) \geq -2\frac{2 + E^0}{\sigma_M + \sigma_m},\quad (11)$$

or, equivalently, for the ratio of the biexciton binding energy E_{XX} with the exciton binding energy $E_X = 2/(\sigma_M + \sigma_m)$,

$$\frac{E_{XX}}{E_X} \geq -(2 + E^0) = 0.1928(1).\quad (12)$$

This lower bound to E_{XX}/E_X , derived only by means of the scaling laws of the Coulomb potential and of the Laplacian, is rather crude. However, it improves the results for the lower bounds for 2D biexciton molecules plotted by Kleinman [13], at least for the range $0.2 \leq m/M \leq 1$.

The upper bound to the binding energy of $(M^+M^+m^-m^-)$ systems can be defined exploiting the concavity of $-1/E(m/M)$ [4]. Defining $E(\lambda) = E(m/M)$ as the ground-state energy for a specific system, one can write

$$\frac{-1}{E(m/M)} = \frac{-1}{E(\lambda)} \geq -\frac{\lambda_{\max} - \lambda}{E(\lambda_{\min})(\lambda_{\max} - \lambda_{\min})} - \frac{\lambda - \lambda_{\min}}{E(\lambda_{\max})(\lambda_{\max} - \lambda_{\min})}, \quad (13)$$

where $\lambda_{\min} \leq \lambda \leq \lambda_{\max}$. Using Eq. (13) together with the exact E_X , the upper bound to the ratio E_{XX}/E_X assumes the analytical form

$$E_{XX}/E_X \leq -2 - \frac{1 + \lambda}{2} \left[-\frac{\lambda_{\max} - \lambda}{E(\lambda_{\min})(\lambda_{\max} - \lambda_{\min})} - \frac{\lambda - \lambda_{\min}}{E(\lambda_{\max})(\lambda_{\max} - \lambda_{\min})} \right]^{-1}. \quad (14)$$

Furthermore, due to the concavity of $-1/E$ over the whole range of mass ratios, Eq. (14) gives, for values of λ external to the interval of definition, i.e., $\lambda > \lambda_{\max}$ and $\lambda < \lambda_{\min}$, a lower bound to the same quantity. To use Eq. (14), we need two energy values, the energy of the dipositronium and the energy of another $(M^+M^+m^-m^-)$ system: for example $H_2 \ ^1\Sigma_g^+$ in two dimensions, Bianchi *et al.* [15] computed an energy of $-5.2703(8)$ hartree by DMC calculations in the Born-Oppenheimer approximation. This value for the minimum of the potential well is equivalent to a binding energy of $1.2703(8)$ hartree with respect to the two hydrogen atoms in two dimensions. The upper bound given by Eq. (14) using dipositronium and $H_2 \ ^1\Sigma_g^+$ as reference systems is shown in Fig. 1, together with the lower bound [Eq. (12)]. If other exact results are available for different mass ratios, it is possible to improve the upper bound to the binding energy simply splitting the $0 \leq \lambda \leq 1$ range in subintervals: inside each one of these subintervals a better upper bound to E_{XX} can be given using Eq. (14). In Fig. 1 we also show an improved upper bound computed splitting the interval in three parts: the DMC energies for the two model biexcitons are $-2.9427(3)$ hartree for $M=2$ and $m=1$, and $-3.3428(10)$ hartree for $M=3$ and $m=1$.

Recently, two independent papers have been published on the binding energy of quasi-2D biexcitons [24,25]. In those works the experimental values of the ratio E_{XX}/E_X , that

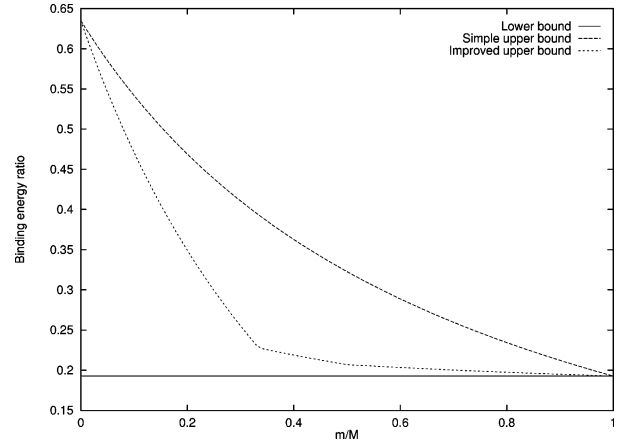


FIG. 1. Upper and lower bounds of the ratio E_{XX}/E_X of $(M^+M^+m^-m^-)$ in two dimensions, plotted vs the mass ratio m/M . This is based on Eqs. (13) and (14), the DMC computed energy of H_2 in two dimensions, and the two model biexciton systems $(M^+m^-)_2$ with $m=1$ and $M=2$ and 3 .

should be independent of the material where the biexcitons are created, were compared with the theoretical results of Kleinman [13]. In both cases the theoretical results were too low, and a model based on a fractional dimension approach was proposed to explain the experimental results. Using this model Birkedal *et al.* [24] found $E_{XX}/E_X = 0.228$, independent of the mass ratio between holes and particles. Since this model assumes a fixed relative geometry for the biexciton, it also introduces some degree of subjectivity and might obscure its dynamical flexibility: this problem can be avoided using models based only on knowledge about the interaction potential between the particles of the system, or computing directly the energies by means of a numerical simulation.

Comparing, for example, the lower bound given by Eq. (12) with the experimental results shown in Refs. [24] and [25], it appears that our result is in optimal agreement with those data, although in our model part of the physics of the system is missing, due to its inability to reproduce the increasing of the binding-energy ratio on going to smaller mass ratios. A better description can be easily obtained by means of the upper bound [Eq. (14)] using the computed values for the energy of biexciton molecules reported previously.

Before discussing which is the best way to deal with these problems, one should note that the experimental results in Refs. [24] and [25] have quite large error bars due to the scarce accuracy of the exciton binding energy E_X . It is our opinion that more accurate measurements of the excitonic binding energy are needed before it becomes possible to discern a meaningful trend in the binding-energy ratio E_{XX}/E_X , and that further theoretical results on these models will prompt the experimentalists to work in this direction.

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