Quantum Monte Carlo results for bipolaron stability in quantum dots

Martin Hohenadler* and Peter B. Littlewood
Theory of Condensed Matter, Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom
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Bipolaron formation in a two-dimensional lattice with harmonic confinement, representing a simplified model for a quantum dot, is investigated by means of quantum Monte Carlo simulations. This method treats all interactions exactly and takes into account quantum lattice fluctuations. Calculations of the bipolaron binding energy reveal that confinement opposes bipolaron formation for weak electron-phonon coupling but abets a bound state at intermediate to strong coupling. Tuning the system from weak to strong confinement gives rise to a small reduction of the minimum Fröhlich coupling parameter for the existence of a bound state.

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INTRODUCTION

Continuous experimental improvements in the preparation and investigation of semiconductor quantum dots in recent years have sparked a lot of interest in extending our theoretical understanding of charge carriers confined in such quasi-zero-dimensional systems.[1] Apart from potential technological applications, quantum dots with tunable properties represent a playground to compare theory and experiment.

The importance of the lattice degrees of freedom in, e.g., polar semiconductors has led to a large number of studies on polarons and bipolarons.[2] These quasiparticles correspond to bound states of a single carrier in a self-induced lattice distortion or of two electrons in a shared virtual phonon cloud. Bipolarons manifest themselves in, e.g., tunneling experiments[3] or transport through molecular dots.[4,5]

Whereas polaron formation in quantum dots is by now quite well understood (see Refs. 6–10 and references therein), conflicting results exist on the stability of bipolarons.[3,8,10–14] These calculations are based on variational treatments, with several works employing strong coupling or adiabatic approximations. It is known from studies of polaron and bipolaron formation that such methods are not able to fully capture the relevant physics,[2,15,16] and their use hence represents a possible source of the contradictory findings. The aim of this paper is to resolve the above issues by applying an unbiased quantum Monte Carlo (QMC) method.[10]

Previous works apply continuum models and the effective mass approximation. Here, we argue that a lattice model is essential for several reasons. First, bipolaron physics involves self-trapping of carriers due to strong interaction with the lattice, a process governed by lattice fluctuations on the scale of the unit cell.[17] Second, for intermediate to strong coupling, bipolarons are rather small, so that any continuum description is expected to break down. Third, quantum dots studied experimentally often contain only a relatively small number of unit cells.

Bipolaron formation in a quantum dot model with local interactions has recently been studied numerically.[10] The purpose of this work is to extend these calculations to a more realistic model with long-range interactions—similar to the continuum models employed by other authors—and to obtain results for the bipolaron binding energy.

MODEL

The Hamiltonian considered here takes the form

$$H = -t\sum_{\langle i,j\rangle,\sigma} c^{\dagger}_{i,\sigma}c_{j,\sigma} + \sum_{i,j} u_{ij}\hat{n}_{i,\uparrow}\hat{n}_{j,\downarrow} + K\sum_{i} |\boldsymbol{r}_i|^2\hat{n}_i + \frac{\omega_0}{2}\sum_{i}(\hat{x}_i^2+\hat{p}_i^2) - \alpha\sum_{i,j} f_{j,i}\hat{n}_i\hat{x}_j, \tag{1}$$

with the long-range Coulomb interaction

$$u_{ij} = \begin{cases} U, & \boldsymbol{r}_i = \boldsymbol{r}_j \\ U/|\boldsymbol{r}_i - \boldsymbol{r}_j|, & \boldsymbol{r}_i \neq \boldsymbol{r}_j \end{cases} \tag{2}$$

and the interaction between an electron at $\boldsymbol{r}_i$ and the oscillator at $\boldsymbol{r}_j$,

$$f_{j,i} = \frac{1}{(|\boldsymbol{r}_j - \boldsymbol{r}_i|^2+1)^{3/2}}. \tag{3}$$

In Eq. (1), $c^{\dagger}_{i,\sigma}$ creates an electron with spin σ at lattice site i (located at $\boldsymbol{r}_i$), $\hat{x}_i$ ($\hat{p}_i$) denotes the displacement (momentum) of the harmonic oscillator at site i, $\hat{n}_{i,\sigma}=c^{\dagger}_{i,\sigma}c_{i,\sigma}$, and $\hat{n}_i=\Sigma_\sigma\hat{n}_{i,\sigma}$. The model parameters are the nearest-neighbor hopping integral t, the Coulomb repulsion U, the confinement strength K, the (dispersionless) optical phonon frequency ω_0, and the electron-phonon coupling constant α. We analyze a two-dimensional (2D) square lattice with two electrons of opposite spins and periodic boundary conditions.

For $|\boldsymbol{r}|\geqslant 1$, the Coulomb interaction u_{ij} in Eq. (2) is of the same form as in previous studies of bipolarons in quantum dots.[8,11,12,14] An important difference between previous work and our lattice model is that u_{ij} does not diverge for $\boldsymbol{r}_i=\boldsymbol{r}_j$. Instead, there is a finite Hubbard $U>0$ for two electrons at the same site, which we believe to be more appropriate for a discussion of small-bipolaron states.

The electron-phonon interaction [Eq. (3)] can be regarded as a lattice version of the Fröhlich model.[18–20] The Holstein-Hubbard model[10] is recovered by setting $u_{ij}=U\delta_{ij}$ and $f_{j,i}=\delta_{ij}$. We define[19,20] $\lambda=2\tilde{E_P}/W$ and $\tilde{E_P}=E_P\Sigma_j f^2_{j,0}$, with the atomic-limit polaron binding energy of the Holstein-Hubbard model, $E_P=\alpha^2/2\omega_0$, and the free bandwidth $W=8t$. In the sequel, all energies will be measured in units of t, and N denotes the lattice size in each dimension. We further set the lattice constant to 1. The 2D case models a disk-shaped

quantum dot with negligible thickness. The dependence on dimensionality has been studied for the Holstein-Hubbard model,[10] and is weak for the Fröhlich model.[19]

The lattice Hamiltonian [Eq. (1)] is appropriate in the study of self-trapping, strong electron-phonon coupling, or strong confinement. Compared to other calculations of phonon effects in quantum dots in the framework of the independent boson model,[21] we take into account the finite size of the dot and hence the possibility of electronic hopping. This is essential because (bi)polaron formation is determined by the balance between kinetic, lattice, and Coulomb energies.

The harmonic (parabolic) confinement potential in Eq. (1) is centered around site (0, 0) [the lattice extends from $-(N-1)/2$ to $(N-1)/2$ in each dimension] and is usually assumed to be a good approximation for real quantum dots.[1] The use of optical bulk phonons is sensible as bipolaron formation is dominated by the coupling to such branches,[3] and the details of the phonon spectrum are of minor importance.[8]

The Hamiltonian [Eq. (1)] contains the relevant terms to describe bipolaron formation in quantum dots. Although more general cases can also be treated with the present method, our lattice model—more amenable to exact numerical treatments than the continuum version—is chosen to be as similar as possible to the models in existing work on bipolaron formation in order to resolve the conflicts in available results.

Finally, due to the simplicity of our model, we refrain from fitting our results to experiment by tuning parameters. Instead, we chose the adiabaticity ratio $\gamma=\omega_0/t=0.1$ (i.e., in the experimentally important adiabatic regime $\gamma \ll 1$) and $U/t=4$, as in previous work,[10,16,22] and vary the electron-phonon coupling and the confinement strength. Note that within the Fröhlich model, U and α are not independent parameters.[18]

METHOD

The worldline QMC method[10,23,24] can be extended to the case of long-range electron-electron interaction[24,26] and an electron-phonon coupling of the Fröhlich type.[19,25] Alternative QMC schemes have also been used to study bipolaron formation.[16,27]

Apart from the controlled (and small) Trotter error, no approximations are made. In particular, the quantum-mechanical nature of the phonon degrees of freedom is fully taken into account, and all interactions are treated on the same footing. For the calculations, we have used a low temperature $\beta t=t/(k_B T)=15$, a sufficiently small Trotter parameter $\Delta\tau=0.05$, and a linear lattice size $N=31$, ensuring small finite-size effects. As pointed out before,[10,28] autocorrelations can be large and must therefore not be ignored.

The bipolaron binding energy is defined as

$$E_B = E(2) - 2E(1), \tag{4}$$

where $E(N_e)$ denotes the total energy of the system with N_e electrons.[31] Krishna *et al.*[14] introduced an additional estimated Coulomb correlation term of two unbound polarons in a quantum dot, which generally leads to enhanced binding energies. Another criterion for the stability of a bipolaron based on the relative distance of the two electrons has also been suggested,[13] yielding a broader region of existence. Definition (4) has been used by most previous authors, and permits comparison to the case $K=0$. Furthermore, E_B, as defined by Eq. (4), has a direct physical meaning as it enters the Boltzmann factor that controls the average number of doubly occupied quantum dots in an ensemble of dots.[8] Note that we use the same exact method to calculate $E(1)$ and $E(2)$.

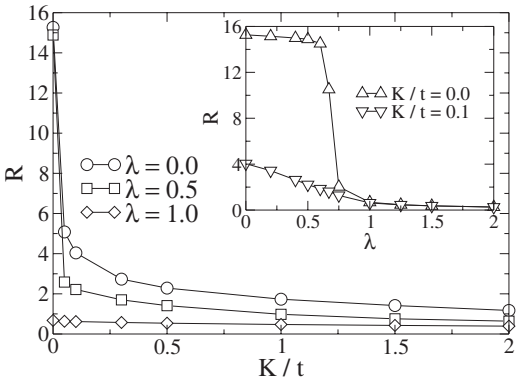

FIG. 1. Average electron-electron distance R as a function of confinement strength K for different values of the electron-phonon coupling parameter λ. The inset shows R as a function of λ for two values of K. Here, $\gamma=0.1$, $U/t=4$, $\beta t=15$ ($\Delta\tau=0.05$), and $N=31$. Lines are guides to the eye, and error bars are smaller than the symbols.

We also measure the average electron-electron separation,

$$R = \left\langle \sum_{i,j} (i-j)^2 \hat{n}_{i,\uparrow} \hat{n}_{j,\downarrow} \right\rangle^{1/2}. \tag{5}$$

For $\lambda=0$, R gives an estimate of the size of the quantum dot, whereas for $\lambda>0$, it may be regarded as a measure for the bipolaron radius. For the calculation of other observables, we refer to a previous paper.[10]

RESULTS

A bipolaron is defined as a bound state of two electrons coupled to the lattice. The energy gain as compared to a system with two noninteracting polarons originates in the additional potential energy from sharing a common lattice polarization cloud. In the atomic limit $t=0$, the latter increases quadratically with the number of electrons per site. This leads to a tendency of the electrons to share the same region of space (i.e., to real-space pairing) and gives rise to a bound state for any $\lambda>0$ if $U=0$ or for $\lambda>\lambda_c(U)$ in the case $U>0$ where the electron-electron Coulomb repulsion has to be overcome.[16,27]

Depending on the model and the parameters, the average distance between the particles can be larger than 1 (large bipolaron), about 1 (intersite bipolaron), or less than 1 (small or onsite bipolaron).[16] For $K=0$, the crossover from a state with two unbound polarons to a bipolaron with increasing λ

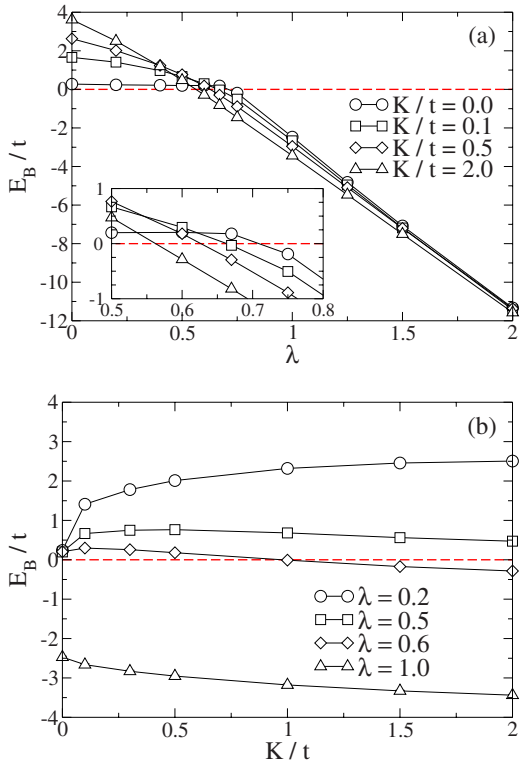

FIG. 2. (Color online) Bipolaron binding energy (a) E_B as a function of λ for different K and (b) E_B as a function of K for different λ. The dashed horizontal line indicates $E_B=0$, and the inset in (a) shows a closeup view.

can be detected from observables such as electron-electron correlation functions or the "radius" R defined by Eq. (5) (see inset of Fig. 1). However, in a confined system, the two-particle wave function is "squeezed" even for $\lambda=0$,[10,13] and the binding energy is the only reliable indicator, with $E_B<0$ for a bound state.

In some previous work on the continuum model, the strength of the confinement potential was measured in terms of a confinement length. Therefore, we start by analyzing the average electron-electron separation as a function of K. Figure 1 shows results for three different values of λ. For $\lambda=0$, the distance rapidly decreases from $R\approx N/2$ at $K=0$ to $R\approx 2-5$ at finite K. For intermediate coupling $\lambda=0.5$, the dependence is similar, but R is systematically smaller than for $\lambda=0$ due to the bipolaron effect. Finally, for strong coupling $\lambda=1$, a small bipolaron is the ground state even for $K=0$, so that confinement has very little influence on R. The range of R in Fig. 1 is comparable to the confinement lengths studied by other authors,[14] and the different behavior for weak and strong couplings is consistent with continuum calculations.[8]

The direct contribution of the confinement term in Eq. (1) to the total energy is linear in N_e and, therefore, cancels when calculating E_B. Consequently, any effect of K on E_B is indirect, mediated by changes in the interaction energies related to lattice distortions and Coulomb correlation.

In Fig. 2(a), we plot the bipolaron binding energy E_B as a function of λ for different values of K. As expected, all curves are strictly monotonic, decreasing with increasing electron-phonon coupling. The binding energy is positive (unbound state) for $\lambda<\lambda_c$ and negative for $\lambda\gtrsim\lambda_c$ (bound state or bipolaron). The inset in Fig. 2(a) reveals a reduction of λ_c with increasing K by about 10%, and the $K=0$ critical coupling is similar to that of the Holstein-Hubbard model [cf. Fig. 7(a) in Ref. 10]. Of course λ_c depends on U, with $\lambda_c\to 0$ as $U\to 0$. For strong coupling, all curves in Fig. 2(a) eventually collapse due to the formation of a small-bipolaron state which is rather insensitive to confinement.

From Fig. 2(a), we can identify three different regimes, for which we plot the binding energy as a function of K in Fig. 2(b). In the regime $\lambda\ll\lambda_c$, the Coulomb interaction is stronger than the phonon-mediated electron-electron attraction, so that we have two polarons with $E_B>0$. Confinement enhances both interactions, and our results reveal that the Coulomb energy becomes even more dominant; i.e., E_B increases with increasing K for $K/t\lesssim 1$. This is particularly evident for the case $\lambda=0$ shown in Fig. 2(a), where E_B increases noticeably with K, approaching the value of the onsite Coulomb repulsion $U/t=4$.

For $\lambda\gg\lambda_c$, a bound state exists as a result of the lattice deformation energy winning over the Coulomb repulsion, and confinement acts in favor of bipolaron binding by further increasing $|E_B|$. The influence of K on the results is much weaker than that for $\lambda\ll\lambda_c$ because the physics is dominated by local correlations, as reflected by the small bipolaron radius (see inset of Fig. 1). The weak dependence of λ_c on K may be attributed to the fact that confinement increases both Coulomb repulsion as well as the phonon-mediated attractive interaction, and thus leaves the effective interaction and λ_c almost unchanged.

In between these two limits, for intermediate λ, E_B shows a nonmonotonic behavior as a function of K. For intermediate coupling, we find a maximum in E_B at small values of K/t and a decrease for an even stronger confinement. Whereas for $\lambda=0.5$ $E_B>0$ for all K shown, a bound state arises for a strong-enough confinement $K/t\gtrsim 1$ and $\lambda=0.6$. In both cases, the variation of E_B with K is rather small. For large K, the curves in Fig. 2(b) approach the corresponding values of the effective onsite interaction $U-2\tilde{E}_P$, which plays an important role in molecular quantum dots with only a single electronic level.[4,5]

The main findings of previous calculations for the continuum Fröhlich model are as follows.[8,10,13] There exists a minimal $\lambda_c(U)$ below which a bound state is suppressed due to Coulomb repulsion. Moreover, some authors argue that there is also a critical confinement strength beyond which the diverging Coulomb repulsion suppresses a bound state for any finite electron-phonon coupling strength $\lambda<\infty$.[11,12,14] No unanimous conclusion has been reached concerning the effect of (weak to intermediate) confinement on the size of the parameter region of existence for the bound bipolaron state.[14]

Let us relate these findings to our results. Similar to previous work and models without confinement, we find a critical coupling λ_c for the formation of a bound state. As pointed out before, within our lattice model, λ_c changes slightly as a function of the confinement strength K and scales approximately linearly with U.[10]

Concerning the regime of strong confinement, our choice of a finite Coulomb repulsion of two electrons with opposite spins located at the same lattice site leads to substantially different physics. Even for $K\to\infty$, a bound onsite bipolaron is formed if the effective onsite interaction $U-2\tilde{E}_P<0$. Finally, we observe that confinement opposes binding for weak coupling $\lambda\ll\lambda_c$, whereas it enhances binding for intermediate to strong coupling $\lambda\gtrsim\lambda_c$. Despite the associated changes of the value of the binding energy, the phase diagram (i.e., the region of existence of a bound state) is only weakly affected by the confinement potential. This insensitivity is even more pronounced for the Fröhlich parameter $\alpha_c\propto\sqrt{\lambda_c}$, which decreases with increasing $K/t\in[0,2]$ by about 5%.

Previous authors have used material-specific parameters to make predictions for the existence of bipolarons in typical quantum dot systems.[14] As we believe that our model is too simple to make quantitative statements, we restrict ourselves to a mostly qualitative discussion. The (small) reduction of the critical coupling due to confinement suggests that the dot size may determine whether bipolarons are stable or not. However, according to our findings, this should only be relevant for materials with intermediate electron-phonon coupling. In contrast, for weak coupling, bipolaron formation is suppressed by Coulomb repulsion, whereas for strong coupling a bipolaron ground state is stable regardless of the confinement strength. Besides, as demonstrated in Fig. 2(b), the value of the binding energy does have a noticeable dependence on the confinement potential, at least for experimentally relevant weak to intermediate values of the electron-phonon coupling. The dot size may also change the phonon spectrum or electronic band structure, but such effects have been neglected in our model.

Finally, we can estimate the order of magnitude of the bipolaron binding energy in real systems by assuming a typical bandwidth of 1 eV. From Fig. 2, we conclude that for plausible values of $0.5\lesssim\lambda\lesssim1$, E_B is a fraction of an eV for $U=0.5$ eV, a value falling into the narrow-band regime $U\gg t$. The binding energy increases with decreasing U, but the physics of bipolaron formation remains qualitatively the same. Our values of E_B are larger than previous variational 3D results.[14] Experimentally, apart from pair tunneling,[3,5] the bipolaron effect should also manifest itself in shot-noise measurements.[29,30]

In summary, we have presented unbiased quantum Monte Carlo results for bipolaron formation in a two-dimensional quantum dot, taking into account the crystal lattice and quantum phonon effects. Confinement is found to give rise to—respectively strengthen—a bound state at intermediate to strong electron-phonon interaction and to reduce the critical coupling for bipolaron formation. The present method can be used to study more general models with dispersive phonons or more complicated electronic bands and dot geometries.[10,25]

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*mh507@cam.ac.uk

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