Interacting-Holstein and extended-Holstein bipolarons

Monodeep Chakraborty,[1,2,*] Masaki Tezuka,[3,†] and B. I. Min[1,‡]

[1]*Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Korea*

[2]*Department of Solid State Physics, Indian Association for the Cultivation of Science, Jadavpur, Kolkata 700032, India*

[3]*Department of Physics, Kyoto University, Kyoto 606-8502, Japan*

(Received 19 June 2013; revised manuscript received 12 January 2014; published 30 January 2014)

Employing the recently developed self-consistent variational basis generation scheme, we have investigated the bipolaron-bipolaron interaction within the purview of the Holstein-Hubbard and the extended-Holstein-Hubbard (F2H) models on a discrete one-dimensional lattice. The density-matrix renormalization group method has also been used for the Holstein-Hubbard model. We have shown that there exists no bipolaron-bipolaron attraction in the Holstein-Hubbard model. In contrast, we have obtained clear-cut bipolaron-bipolaron attraction in the F2H model. Composite bipolarons are formed above a critical electron-phonon coupling strength, which can survive the finite Hubbard U effect. We have constructed the phase diagram of F2H polarons and bipolarons, and discussed the phase separation in terms of the formation of composite bipolarons.

DOI: 10.1103/PhysRevB.89.035146 PACS number(s): 74.50.+r, 74.20.Rp, 72.25.−b, 74.70.Tx

I. INTRODUCTION

Bipolarons play an important role in many areas of material science and biological science [1–3]. Many-body techniques such as the density-matrix renormalization group (DMRG) and the quantum Monte Carlo (QMC) have been employed successfully to investigate bipolarons in Holstein and Fröhlich systems at half-filling or quarter-filling [2,4–9]. The continuum Fröhlich bipolarons were also investigated [10–13]. On the other hand, a proper description of bipolaron-bipolaron (b-b) interaction in the dilute limit is also very important, as it gives rich information from an entirely different perspective. However, studies on the b-b interaction in the dilute limit are still lacking. The primary reason is that variational approaches based on the exact diagonalization (VAED), which have been very successful for polaron and bipolaron problems [14–20], cannot be applied to even four-electron (two bipolarons) problems. Chakraborty *et al.* [21] has recently developed a new self-consistent scheme of generating variational basis based on the exact diagonalization (SC-VAED), which makes the b-b interaction problem tractable in the dilute limit.

In this paper, we have applied the SC-VAED scheme to Holstein-Hubbard and extended-Holstein-Hubbard (F2H) [19] systems with four and six electrons. We have also employed the DMRG method to a four-electron Holstein system to check the results of the SC-VAED scheme. Our calculations reveal that the Holstein type of electron-phonon (e-ph) interaction does not yield the attractive interaction between two bipolarons. In contrast, the e-ph interaction of extended-Holstein F2 type produces the b-b attractive interaction above a certain e-ph coupling to form composite bipolarons. Hereafter, we name these composite bipolarons as b *composite*. The on-site Hubbard electron-electron (e-e) interaction U, when it is small, weakens the interbipolaron binding as well as the intrabipolaron binding [15,19], producing a lighter b composite. An increase in U leads to the dissociation of the b composite into two repelling bipolarons, and then into individual polarons. However, above a critical e-ph coupling, both the interbipolaron and intrabipolaron bindings survive infinite U. We have constructed the phase diagram of F2H polarons and bipolarons with respect to e-ph and e-e interactions, and discussed the phase separation phenomenon in terms of the b-composite formation.

This paper is organized as follows. In Sec. II, we introduce the Hamiltonian, which incorporates the e-e and e-ph interactions within the one-dimensional (1D) Holstein-Hubbard and F2H models. We also provide computational details here. In Sec. III, we have examined the bipolaron binding for the four-electron Holstein-Hubbard model, using the SC-VAED and the DMRG methods. In Sec. IV, we have made a detailed analysis of the four-electron F2H model on the basis of calculated binding energies and correlation functions in different parameter regimes. We have also discussed the results of the six-electron system within the F2 model. Then we have constructed the phase diagram of bipolarons with respect to e-ph interaction strength λ and the on-site Hubbard U. Conclusions follow in Sec. V.

II. HAMILTONIAN AND COMPUTATIONAL DETAILS

The Fröhlich-Hubbard Hamiltonian on a discrete 1D lattice [22,23] is defined as follows:

$$H = -\sum_{i,\sigma}(tc^{\dagger}_{i,\sigma}c_{i+1,\sigma} + \text{H.c.}) + \omega\sum_{j}a^{\dagger}_{j}a_{j} + g\omega\sum_{i,j,\sigma}f_{j}(i)n_{i,\sigma}(a^{\dagger}_{j} + a_{j}) + U\sum_{i}n_{i,\uparrow}n_{i,\downarrow}, \quad (1)$$

where $c^{\dagger}_{i,\sigma}$ ($c_{i,\sigma}$) creates (annihilates) an electron of spin σ at site i and $a^{\dagger}_{j}$ (a_{j}) creates (annihilates) a phonon at site j. We take the spin-$\frac{1}{2}$ electron ($S_z = +\frac{1}{2}$ or $-\frac{1}{2}$). The third term represents the coupling of an electron at site i with an ion at site j with g being the dimensionless e-ph coupling parameter. $f_j(i)$ is the long-range e-ph interaction, the actual

*Corresponding author. sspmc2@iacs.res.in

†tezuka@scphys.kyoto-u.ac.jp

‡bimin@postech.ac.kr

form of which is given by [23]

$$f_j(i) = \frac{1}{(|i-j|^2+1)^{3/2}}. \quad (2)$$

The *e-ph* coupling strength λ is defined by [15,23]

$$\lambda = \frac{\omega g^2 \sum_l f_l^2(0)}{2t}. \quad (3)$$

If $f_j(i) = 0$ for $i \neq j$, the Fröhlich-Hubbard model becomes the Holstein-Hubbard model. In our study, the Fröhlich *e-ph* interaction is approximated by the extended-Holstein interaction of F2 type, which corresponds to the case of $f_{i\pm(1/2)}(i) = 1$ and zero otherwise, as defined by Bonča and Trugman [15] and Chakraborty *et al.* [19]. The F2 model couples an electron with two nearest-neighbor ions placed in the interstitial [15,19]. We set the electron hopping $t = 1$ for numerical calculations, and so all energy parameters are expressed in units of t.

The variational basis is generated starting with a state of bare electrons and adding new states by repeated application of the Hamiltonian [14,16,18–20]. The off-diagonal terms of the Hamiltonian generate different configurations of phonons and electrons for a specific up-spin electron ($\uparrow$) at position i [14].

All translations of these states on the periodic 1D lattice are included. In the SC-VAED scheme, we first generate a relatively small basis set and obtain the ground state energy and the wave function. Then the states with highest probability are identified, and a new basis is generated by application of the Hamiltonian on these chosen highly probable states. Accordingly the size of the basis is increased. The ground state energy and the wave function are calculated again. This process is continued in a self-consistent way, by increasing the basis size at each cycle until the desired accuracy in the ground state energy is obtained [21]. At each step, the weight of m-phonon states for the ground state, $|C_0^m|^2$, as defined by Fehske *et al.* [23] is calculated. The convergence of $|C_0^m|^2$ is checked to ensure that the basis contains an adequate number of phonons required at the given parameter regime. We have considered lattice sizes up to $L = 24$ for the four-electron case and $L = 12$ for the six-electron case. In principle, for the four-electron case, we can go beyond $L = 24$, probably up to $L = 32$, but that would take much more computation time [24]. For the six-electron case, we can go up to $L = 18$, but again that would take much more computation time.

With obtained ground state energies and wave functions, we discuss the *b-b* interaction in terms of the binding energy (Δ) and the *e-e* correlation function [$C_{\uparrow,\sigma}(i-j)$]. Δ for four- and six-electron systems are defined by [14,19]

$$\Delta_4 = E_4 - 2 \times E_2, \quad (4)$$

$$\Delta_6 = E_6 - E_4 - E_2, \quad (5)$$

where E_2, E_4, and E_6 are ground state energies of two-electron (one up-spin electron and one down-spin electron), four-electron (two up-spin electrons and two down-spin electrons), and six-electron (three up-spin electrons and three down-spin electrons) systems, respectively. $C_{\uparrow,\sigma}(i-j)$ is defined by the probability of finding the other electron(s) with respect to one up-spin electron, at the position of which the basis is generated [14,19]:

$$C_{\uparrow,\sigma}(i-j) = \langle \Psi_0 | n_{i,\uparrow} n_{j,\sigma} | \Psi_0 \rangle, \quad (6)$$

where $n_{j,\sigma}$ is the number operator with spin σ at site j. In Eq. (6), when $i = j$, $\sigma = \uparrow$ is not considered due to the Pauli exclusion principle. $|\Psi_0\rangle$ is the ground state wave function. Note that, in this work, we have always considered the same numbers of up-spin and down-spin electrons and limited the basis states in SC-VAED to ones with total spin $S = 0$.

TABLE I. Ground state energies of the four-electron Holstein lattice (E_4) for different numbers of lattice sites obtained using the SC-VAED and the DMRG method. $\omega = 1.0$ and $\lambda = 0.5$ are fixed. Converged energy is still higher than twice the ground state energy of the Holstein bipolaron, -10.8493. Energies are in units of t.

Sites	E_4 (SC-VAED)	E_4 (DMRG)
12	-10.77	-10.782
16	-10.81	-10.797
24	-10.83	-10.828
60		-10.846

III. HOLSTEIN-HUBBARD MODEL

We have calculated ground state energies of four-electron (two up-spin and two down-spin electrons) Holstein lattices using both the SC-VAED and the DMRG method. The former was done in the periodic boundary condition, while the latter [25] was done in the open-boundary condition. Results of both methods for different ω and λ do not show any signature of binding of the two bipolarons, i.e., two on-site *spin-antiparallel* ($S = 0$) bipolarons repel each other. For all parameter regimes, the ground state energies of the four-electron Holstein system are higher than twice the ground state energy of individual Holstein bipolarons.

Let us look at the obtained ground state energies and correlation functions for $\omega = 1.0$, $\lambda = 0.5$ as a typical example. Twice the ground state energy of a Holstein bipolaron at this regime is -10.8493 and Table I shows the calculated energies for the four-electron system at this parameter regime for different lattice sizes. Note that the converged energy is higher than twice the ground state energy of the Holstein bipolaron. This result is quite expected because an intersite spin-parallel ($S = 1$) bipolaron, which is formed of two nearest-neighbor electrons with the same spin, is not favored within the Holstein paradigm [15,19]. The spin-antiparallel bond helps formation of two bipolarons by binding up-spin and down-spin electrons (hence two spin-antiparallel bipolarons are formed from two up-spin and two down-spin electrons). However, the absence of a spin-parallel bond in the Holstein model obliterates the possibility of two bipolarons gluing together.

Figure 1 provides the *e-e* correlation function $C_{\uparrow,\sigma}(i-j)$ for the ground state of the four-electron Holstein system at $\lambda = 0.5$ and $\omega = 1.0$. $C_{\uparrow,\sigma}(i-j)$'s calculated for three different system sizes (12-, 16-, and 24-site) show that the two bipolarons tend to maintain the maximum separation as far as the system size allows. It should be noted, that $\pm L/2$ for different system sizes are the same point. Hence, the sum rule is satisfied for $-L/2+1$ to $L/2$ (to avoid double counting for

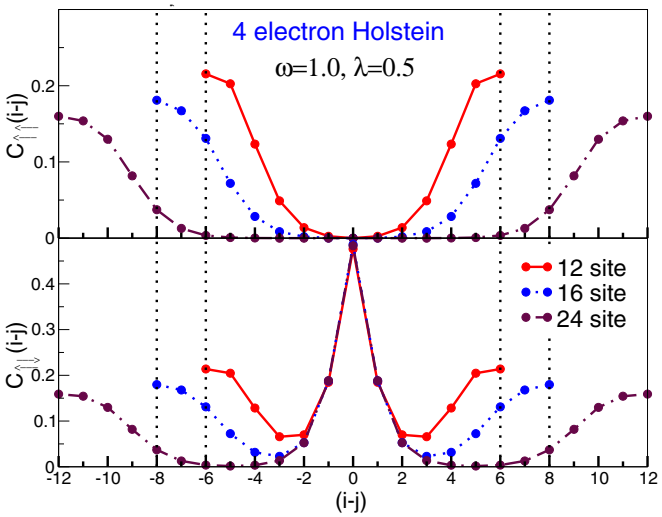

FIG. 1. (Color online) The *e-e* correlation function $C_{\uparrow,\sigma}(i-j)$ for the four-electron Holstein lattice ($U=0$). The position i and spin of the electron $\uparrow$ is fixed, since the basis has been generated with respect to it. $C_{\uparrow,\sigma}(i-j)$'s for 12-, 16-, and 24-site Holstein lattices are compared. It should be noted, that $\pm L/2$ for different system sizes are the same point.

the end point). As shown in Table I, the ground state energy of the four-electron system is lowered with increasing the system size, which indicates the repulsive nature of interaction between the two bipolarons. The finite size effect is evident in this case, as two bipolarons want to be far apart from each other, but the finite size of our lattice limits that. Hence, the bigger the lattice size, the closer the ground state energy is to twice the energy of the individual bipolarons. The 60-site result by the DMRG method validates this conjecture further (see Table I). Therefore the formation of a spin-parallel bipolaron is unstable within the Holstein paradigm [15], and the scenario remains the same for the four-electron case too. The on-site Hubbard U has the same effect on the individual bipolarons, as reported earlier [14,19], and would not lead to binding between the two bipolarons.

IV. F2H MODEL

According to earlier calculations [15,19], a longer-ranged *e-ph* interaction facilitates the binding of the spin-parallel bipolaron above a critical *e-ph* coupling strength λ_c^b. Here we have studied the four-electron F2H lattice [15,19], using the SC-VAED method. First, we have considered the case of $U=0$. Figure 2(a) shows $C_{\uparrow,\sigma}(i-j)$ for $\lambda=0.1$ at $\omega=1.0$. In this case, we do not obtain *b-b* binding. Indeed, as shown in Table II, converged ground state energy of the four-electron F2 lattice for $\lambda=0.1$, -8.6024 ($L=24$), is higher than twice the ground state energy of the F2 bipolaron, -8.6215. On the contrary, at larger $\lambda=1.0$ in Fig. 2(b), $C_{\uparrow,\sigma}(i-j)$ clearly demonstrates the formation of a strongly bound b composite, which is seen to be valid independently of the system size. In this case, converged ground state energy is -24.5293 ($L=24$), which is lower than twice the ground state energy of the F2 bipolaron, -18.3669.

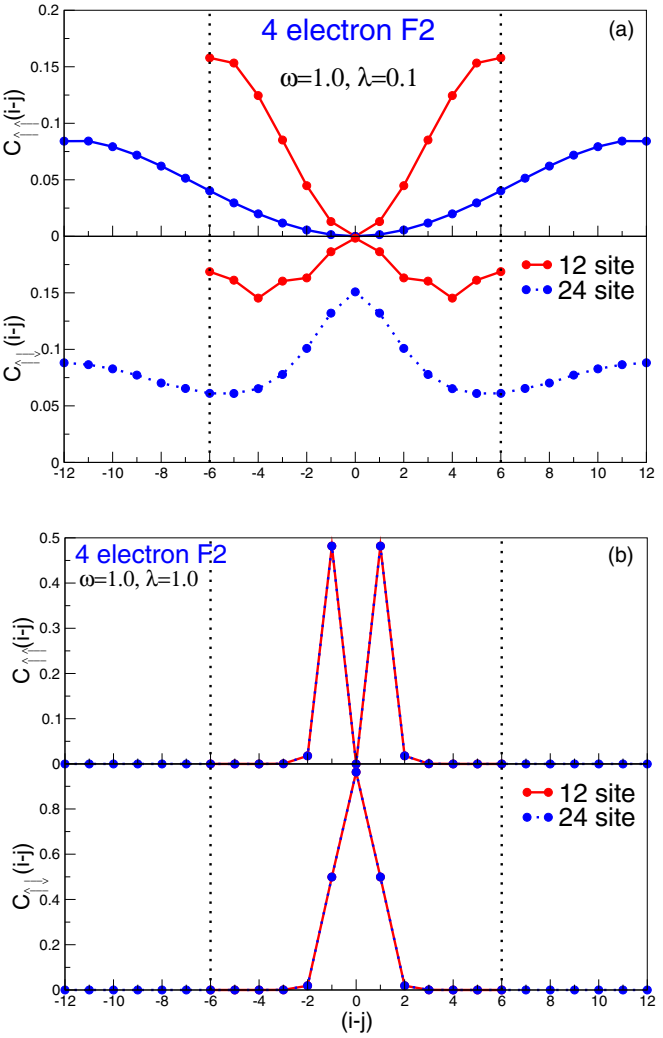

FIG. 2. (Color online) The *e-e* correlation function $C_{\uparrow,\sigma}(i-j)$ for the four-electron F2 lattice. Two different system sizes (12 and 24) are considered. (a) $\omega=1.0$, $\lambda=0.1$ and (b) $\omega=1.0$, $\lambda=1.0$. It should be noted, that $\pm L/2$ for different system sizes are the same point.

Figure 3 shows the binding energy Δ_4 of the four-electron b composite as a function of λ. We see no *b-b* binding for small λ, while above $\lambda_c^{b-b}\approx 0.3$, Δ_4 becomes negative, signaling the formation of b composite. It is notable that, for large λ, Δ_4 becomes almost independent, at least up to two decimal places, of the system size (see Table II). The inset of Fig. 3 shows Δ_4's for smaller system sizes. For small systems of $L=4-8$, b composite is apparently formed even for $\lambda<\lambda_c^{b-b}$. This is attributed to the finite size effect. With the increase in λ, the polaronic nature of the individual bipolaron increases, and due to the small size of the lattice, their wave functions are forced to overlap to exhibit a spurious binding.

Table II presents the ground state energies of four-electron F2 lattices (E_4) for different system sizes and λ. Bipolaron (two-electron) ground state energies (E_2) at the same parameters are also provided for comparison. At $\lambda=0.1$, as the system size increases, the ground state energy approaches twice the bipolaron energy, -8.6215. This suggests that the interaction between the two bipolarons is repulsive. An

TABLE II. Ground state energies of four-electron F2 lattices (E_4) at different λ for different system sizes (L) ($\omega = 1.0$ is fixed). Twice the F2 bipolaron energies are also given in the last column. Energy parameters are in units of t.

	E_4						$2 \times E_2$
λ	$L = 4$	$L = 8$	$L = 12$	$L = 16$	$L = 20$	$L = 24$	$L = 39$
0.1	−7.3136	−8.3763	−8.5402	−8.5831	−8.5996	−8.6024	−8.62147
0.2	−8.9795	−9.3858	−9.4028	−9.3840	−9.3835	−9.3776	−9.41269
0.3	−10.6475	−10.4364	−10.3432	−10.2867	−10.2917	−10.2698	−10.31085
0.4	−12.3300	−11.6105	−11.5521	−11.5469	−11.5586	−11.5578	−11.28980
0.5	−14.0364	−13.2887	−13.2831	−13.2876	−13.2882	−13.2902	−12.33584
0.6	−15.8017	−15.3675	−15.3668	−15.3714	−15.3715	−15.3722	−13.44075
0.7	−17.7584	−17.5801	−17.5891	−17.5956	−17.5956	−17.5960	−14.59928
0.8	−19.9535	−19.8156	−19.8709	−19.8791	−19.8792	−19.8793	−15.80812
0.9	−22.2382	−22.0267	−22.1811	−22.1942	−22.1944	−22.1945	−17.06493
1.0	−24.5580	−24.5270	−24.5292	−24.5279	−24.5292	−24.5293	−18.36692

increase in λ renders more polaronic nature to these repulsive bipolarons, and makes the numerical task more difficult. Note that, above $\lambda_c^{b-b} \approx 0.3$, the binding energy becomes negative for all system sizes, and the ground state energy is converged at least up to two decimal places. The finite size effect is significant in the F2 system as well for small λ, for which there is no b-b binding for the same reason as mentioned for the Holstein system. However, for large λ the finite size effect becomes much smaller because the spatial extent of constituent bipolarons and that of b composite (formed out of coalescing of two bipolarons) are small. Table II amply demonstrates this fact.

The energy behaves nonmonotonically with L for some values of λ in Table II. For $\lambda \lesssim 0.3$, the interaction between bipolarons is repulsive and the ground state energy of the system with four fermions should converge to twice the ground state energy of the system with two fermions (bipolaron); Δ should converge to zero as $L \to \infty$.

As λ increases from zero, our numerical calculation suffers from two types of difficulties:

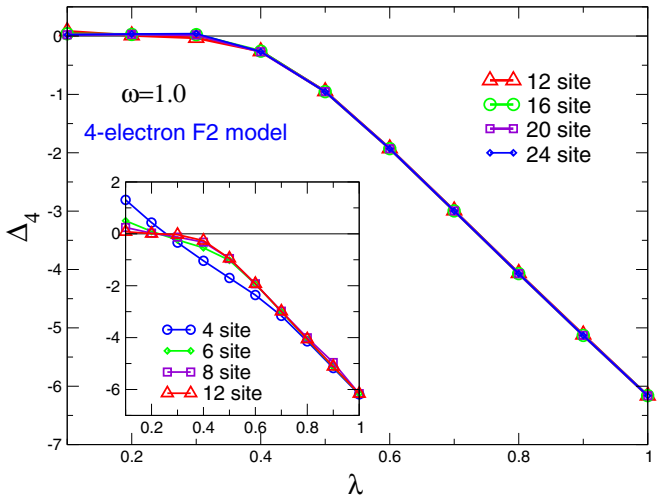

FIG. 3. (Color online) The binding energy (Δ_4) of the four-electron F2 b composite as a function of the e-ph coupling strength λ for different system sizes. The inset shows Δ_4's for small system sizes. $\omega = 1.0$, $U = 0$, and energies are in units of t.

(a) Because of the finite extent (rather large) of the bipolaron, two bipolarons can interact with each other along the opposite path as well as along the confronted path, when the system size is small under periodic boundary condition. This effect can produce a false binding.

(b) When two bipolarons are actually not binding, a significantly larger basis size is required to get the same accuracy when the system size increases. Therefore, within a realistic computational effort, it remains very difficult to obtain a universal scaling law of the value of Δ with respect to L.

On the other hand, for $\lambda \geqslant 0.4$ where the bipolarons attract each other, although we observe an overestimation of Δ for very small lattice sizes, the addition of lattice sites in the periodic boundary condition does not significantly increase the numerical complexity. In other words, the effect we have described above is less significant and consequently we can observe that electron-electron correlation function for $\lambda = 1.0$ shown in Fig. 2(b) shows a very good convergence between $L = 12$ and $L = 24$ and so do their corresponding ground state energies. For values of λ at which we get a bipolaron composite, the states with phonons and electrons far away from the composite center contribute insignificantly to the ground state, and Δ (which is negative) would show a convergence similar to that of the ground state energies. This situation is the same as in the bipolaron (bound state of two polarons), where the ground state energy (and hence the Δ) converges up to seven decimal places with $L = 37$ and remains the same even for higher L [15,19].

Bonča and Trugman [15] showed through their analytical and numerical considerations that the F2 spin-parallel ($S = 1$) bipolaron is stabilized above $\lambda_c^b = 0.76$ at $\omega = 1.0$. On the other hand, we have found, that in the above case of a four-electron system, the two bipolarons bind above $\lambda_c^{b-b} = 0.3$, which is much lower than $\lambda_c^b = 0.76$. The reason why the b-b binding takes place at lower λ is due to the presence of an intersite ($S1$) spin-antiparallel bond between the two adjacent on-site ($S0$) bipolarons which stabilizes the spin-parallel bond between them.

We substantiate this behavior with our six-electron F2 lattice calculations. Figure 4 shows Δ_6's of the six-electron F2 system as a function of λ. It is seen that a b composite of three bipolarons is to be formed for $\lambda \gtrsim 0.3$. The numerical

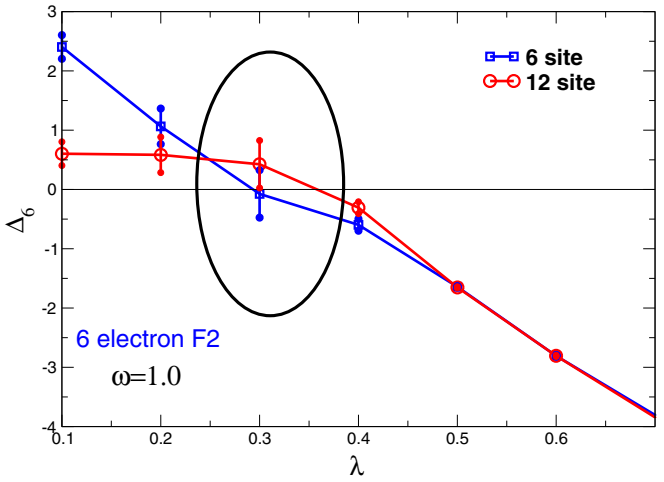

FIG. 4. (Color online) Binding energies for six-electron F2 lattices (Δ_6's) as a function of λ ($\omega = 1.0$ and $U = 0$ are fixed). Two system sizes ($L = 6$ and $L = 12$) are considered. The numerical precision is rather low in the region marked by the ellipse. Energies are in units of t.

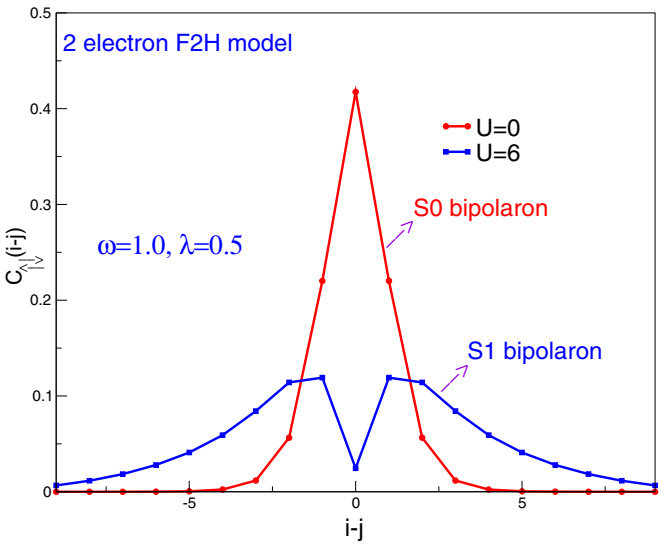

FIG. 5. (Color online) The e-e correlation function $C_{\uparrow,\downarrow}(i-j)$ for the two-electron F2H lattice. Two values of Hubbard U are considered at $\omega = 1.0$ and $\lambda = 0.5$. The red line with circles represents the formation of an on-site $S0$ bipolaron for $U = 0$, while the blue line with squares represents the formation of an intersite $S1$ bipolaron for $U = 6.0$. The b composite formed of two $S0$ bipolarons would have two spin-parallel bonds, while that formed of two $S1$ bipolarons would have either only one $S1$ spin-parallel bond or only one $S1$ spin-antiparallel bond.

precision is not as high as the four-electron case, and is especially lower for the region marked by the ellipse. Once there occurs a b-b binding, the numerical precision is improved, but at best up to one to two decimal places. Better numerical precision can be obtained, but that would be too expensive (basis states of the order of 2–3 $\times$ 10^7) and time consuming (a few weeks to a month). In any way, the six-electron result qualitatively validates the fact that the bipolarons glue into a b composite above a certain $\lambda \sim 0.3$ (for $\omega = 1.0$).

The on-site Hubbard e-e interaction U has an interesting consequence in the formation of b composite. Due to the on-site nature of U, it is tempting to expect that U does not affect the spin-parallel wave function that is responsible for the b-b binding. However, physics is not so simple. Let us recall the case of spin-antiparallel bipolaron formation for the two-electron case of the F2H model. As shown in Fig. 5, with increasing U, the on-site $S0$ bipolaron formed at $U = 0$ transforms into an intersite $S1$ bipolaron. Now consider two cases of b-b bindings between $S0$ bipolarons and between $S1$ bipolarons. Two $S0$ bipolarons, once they are bound, would produce two spin-antiparallel $S1$ bonds, which stabilizes the two $S1$ spin-parallel bonds. On the other hand, above a finite U, the two resulting neighboring $S1$ bipolarons would have either only one $S1$ spin-antiparallel bond or only one $S1$ spin-parallel bond, which will not be sufficient to stabilize the b-b binding. Therefore, with increasing U, the individual bipolaron transforms from $S0$ to $S1$, which would result in the weakening of b-b binding between the bipolarons. Therefore, on-site U interaction really influences the b-b binding, i.e., with increasing U, the b composite already formed above a certain $\lambda_c^{b\text{-}b}$ would dissociate into individual $S1$ bipolarons, and these individual bipolarons would break up into individual polarons with a further increase in U [15,19].

Figure 6 shows the ground state energies of the F2H polaron, bipolaron, and b composite of four-electron systems as a function of U. We have discussed above that, for $\lambda > \lambda_c^b = 0.76$, an individual spin-parallel bipolaron can be formed. For $\lambda = 0.65$ ($< \lambda_c^b$) in Fig. 6(a), the energy of the four-electron b composite is the lowest at small U. However, with increasing U, it becomes higher than twice the energy of the bipolaron and then even higher than four times the energy of the polaron. In contrast, for $\lambda = 0.85$ ($> \lambda_c^b$) in Fig. 6(b), it

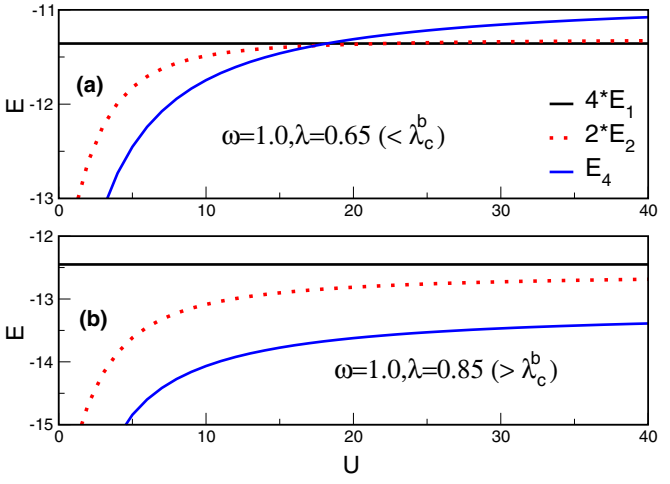

FIG. 6. (Color online) Ground state energies of b composite of four-electron (E_4), bipolaron (E_2), and polaron (E_1) in the F2H lattice ($L = 6$) as a function of U. The blue solid line represents the energy of b composite, the red dotted line represents twice the energy of the bipolaron, and the flat black line represents four times the energy of the polaron. (a) Energies at $\lambda = 0.65$, which is less than $\lambda_c^b = 0.76$. (b) Energies at $\lambda = 0.85$ ($> \lambda_c^b$). Energy parameters are in units of t, and $\omega = 1.0$ is fixed.

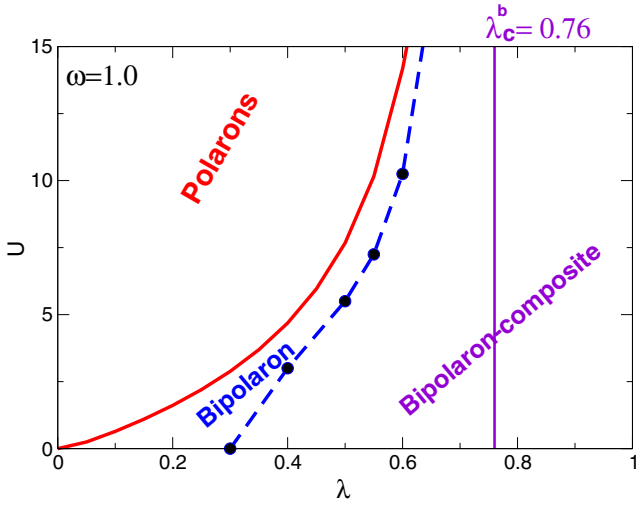

FIG. 7. (Color online) Phase diagram of F2H polarons with respect to *e-ph* coupling strength λ and on-site *e-e* interaction U ($\omega = 1.0$ is fixed). The blue dashed line separating the *b* composite from bipolarons is a qualitative guide for the eye.

is observed that the four-electron *b* composite is always lower in energy than individual bipolarons and polarons. This feature indicates that the *b* composite formed above λ_c^b remains glued even for very large U.

Figure 7 displays the phase diagram of F2H polarons with respect to λ and U. There are three distinct regimes: (i) individual polarons, (ii) individual bipolarons, and (iii) *b* composite. Independent bipolarons at very small λ break into repulsive polarons with increasing U. At larger λ and $U = 0$, repulsive bipolarons coalesce into the *b* composite. With increasing U, this again dissociates into repulsive bipolarons, and then into repulsive polarons with a further increase in U. However, beyond a certain critical $\lambda_c^b = 0.76$, the *b* composite survives the effect of infinite U. Higher U results in lighter *b* composite, as U would significantly bring down the effective mass of the system and make it more mobile.

While the dashed line in Fig. 7 is qualitative in nature because its numerical accuracy is not very high, it should be noted that the phase boundary between the polarons and bipolarons (represented by the red solid line) is quite accurate as it is obtained from the bipolaron (two-electron) calculation [15,19]. Also the vertical line representing the critical λ, above which the stability of *b* composite is unaffected by U, is an analytical result by Bonča and Trugman [15]. The change of curvature of the dashed line is more of a numerical artifact.

We conjecture that the formation of *b* composite and its evolution with U are closely related to phenomena of phase separation observed in various systems, such as colossal magnetoresistance (CMR) manganites and high-T_c superconductors [26,27]. Bonča and Trugman [15] suggested that, for a system with $\lambda > \lambda_c^b$, a third electron (polaron) could stick to already formed bipolaron, which would bring about the phase separation. Recently, Hohenadler *et al.* [8] also showed that the extended *e-ph* interaction yields the phase separation. Our study corroborates this scenario. We have demonstrated through calculations for four-electron and six-electron F2H systems that the longer-range *e-ph* interaction induces the gluing of bipolarons so as to produce a stable *b* composite, which is reminiscent of the phase separation phenomenon. Our study thus sheds light on the microscopic description of phase separation in the presence of both *e-ph* and *e-e* interactions.

V. CONCLUSIONS

We have investigated four- and six-electron Holstein-Hubbard and F2H systems, treating the electron-electron and electron-phonon interactions on an equal footing in the framework of the SC-VAED method. The Holstein type of on-site *e-ph* interaction does not support the formation of *b* composite, as confirmed by both the SC-VAED and the DMRG calculations. In contrast, the F2H type of *e-ph* interaction leads to the formation of *b* composite above a certain *e-ph* coupling strength $\lambda_c^{b\text{-}b} \approx 0.3$ (at $\omega = 1.0$). We have shown that, with increasing Hubbard U, this *b* composite is dissociated into individual bipolarons and then into individual polarons. However, above a critical $\lambda_c^b = 0.76$ (at $\omega = 1.0$), the *b* composite survives the effect of U, forming coalesced $S1$ bipolarons. Our unbiased study of *e-ph* and *e-e* interacting systems would provide insight into the understanding of phase separation physics, which is central to many areas of condensed matter physics.

ACKNOWLEDGMENTS

This work was supported by the NRF (Grant No. 2009-0079947) and the POSTECH Physics BK21 fund. The work of M.T. was partially supported by the Grant-in-Aid for the Global COE Program "The Next Generation of Physics, Spun from Universality and Emergence" from MEXT of Japan. Stimulating discussions with Beom Hyun Kim, Bongjae Kim, and Holger Fehske are greatly appreciated.

[1] M. Hohenadler, M. Aichhorn, and W. von der Linden, Phys. Rev. B **71**, 014302 (2005).
[2] H. Fehske, G. Wellein, G. Hager, A. Weisse, and A. R. Bishop, Phys. Rev. B **69**, 165115 (2004).
[3] Numerical solution of the Holstein polaron problem; H. Fehske and S. A. Trugman, *Polarons in Advanced Materials*, edited by A. S. Alexandrov, Springer Series in Material Sciences Vol. 103 (Springer-Verlag, Dordrecht, 2007), pp. 393–461.
[4] M. Tezuka, R. Arita, and H. Aoki, Phys. Rev. Lett. **95**, 226401 (2005); Phys. Rev. B **76**, 155114 (2007).
[5] R. P. Hardikar and R. T. Clay, Phys. Rev. B **75**, 245103 (2007).
[6] K.-M. Tam, S.-W. Tsai, D. K. Campbell, and A. H. Castro Neto, Phys. Rev. B **75**, 161103 (R) (2007).
[7] H. Fehske, G. Hager, and E. Jeckelmann, Europhys. Lett. **84**, 57001 (2008).
[8] M. Hohenadler, F. F. Assaad, and H. Fehske, Phys. Rev. Lett. **109**, 116407 (2012).

[9] M. Hohenadler and F. F. Assaad, Phys. Rev. B **87**, 075149 (2013).
[10] G. Verbist, F. M. Peeters, and J. T. Devreese, Phys. Rev. B **43**, 2712 (1991).
[11] G. Verbist, M. A. Smondyrev, F. M. Peeters, and J. T. Devreese, Phys. Rev. B **45**, 5262 (1992).
[12] F. Luczak, F. Brosens, and J. T. Devreese, Phys. Rev. B **52**, 12743 (1995).
[13] J. T. Devreese, S. N. Klimin, and V. M. Fomin, Phys. Rev. B **63**, 184307 (2001).
[14] J. Bonča, T. Katrasnik, and S. A. Trugman, Phys. Rev. Lett. **84**, 3153 (2000).
[15] J. Bonča and S. A. Trugman, Phys. Rev. B **64**, 094507 (2001).
[16] J. Bonča, S. A. Trugman, and I. Batistic, Phys. Rev. B **60**, 1633 (1999).
[17] Li-Chung Ku, S. A. Trugman, and J. Bonča, Phys. Rev. B **65**, 174306 (2002).
[18] A. Chakrabarti, M. Chakraborty, and A. Mookerjee, Physica B **388**, 63 (2007).
[19] M. Chakraborty, B. I. Min, A. Chakrabarti, and A. N. Das, Phys. Rev. B **85**, 245127 (2012).
[20] M. Chakraborty, A. N. Das, and A. Chakrabarti, J. Phys.: Condens. Matter **23**, 025601 (2011).
[21] M. Chakraborty and B. I. Min, Phys. Rev. B **88**, 024302 (2013).
[22] A. S. Alexandrov and P. E. Kornilovitch, Phys. Rev. Lett. **82**, 807 (1999).
[23] H. Fehske, J. Loos, and G. Wellein, Phys. Rev. B **61**, 8016 (2000).
[24] For the $L = 24$ case, it takes typically six to seven days CPU time.
[25] The DMRG calculation considered up to 2^5-1 phonons per site and up to 400 density matrix eigenstates retained at each step of the iteration.
[26] A. S. Aleksandrov and V. V. Kabanov, Sov. Phys. Solid State **28**, 631 (1986).
[27] V. B. Shenoy, D. D. Sarma, and C. N. R. Rao, Chem. Phys. Chem. **7**, 2053 (2006).