

Bond bipolarons: Sign-free Monte Carlo approach

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Polarons originating from phonon displacement modulated hopping have relatively light masses and thus are of significant current interest as candidates for the bipolaron mechanism of high-temperature superconductivity [Sous *et al.*, *Phys. Rev. Lett.* **121**, 247001 (2018)]. We observe that the bond model, when the dominant coupling comes from atomic vibrations on lattice bonds, can be solved by efficient sign-free Monte Carlo methods based on the path-integral formulation of the particle sector in combination with either the (real-space) diagrammatic or Fock-path-integral representation of the phonon sector. We introduce the corresponding algorithms and provide illustrative results for singlet bipolarons in two dimensions. The data suggest that the route towards high-temperature superconductivity (if any) in the multiparametric space of the model lies between the Scylla of large size of moderately light bipolarons and the Charybdis of large mass of compact bipolarons. As a result, on-site repulsion is helping *s*-wave superconductivity in sharp contrast with existing expectations.

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Introduction. Broad interest in polarons—stable quasi-particles emerging as a result of strong renormalization of properties of a bare particle due to interaction with this or that environment—comes from their ubiquitous presence across all fields of physics. There exist electron-phonon polarons [1–8], spin polarons [9–11], Fermi polarons [12,13], protons in neutron-rich matter [14], etc. Coupling to the environment may also lead to formation of bound particle states called bipolarons. Their study is motivated by the possibility of identifying a possible mechanism for high-temperature superconductivity when a gas or liquid of bound pairs of fermions undergoes a superfluid (SF) transition at temperature

$$T_c \sim \frac{n^{2/d}}{m_*}, \quad (1)$$

where n is the bipolaron density in a system of dimension $d \geq 2$ and m_* is the bipolaron effective mass. (We assume that effective interactions between the bipolarons are repulsive.) The estimate for T_c is very robust against repulsive interactions: In $d = 3$ it barely changes between the gas and liquid densities [15–19], and in $d = 2$ the dependence on interactions is logarithmically weak [19–22]. Since the SF transition temperature increases with density, the highest value of T_c for this mechanism is obtained when the interparticle distance is of the order of the pair size, or $nR_*^d \sim 1$, where R_* is the bipolaron radius [23,24]

$$T_c \sim \frac{1}{m_* R_*^2}. \quad (2)$$

At higher density, the strongly correlated state of fermions is no longer captured by the bipolaron physics adequately. In principle, it can feature even higher values of T_c

before forming antiferromagnetic and bond-ordered insulating phases [25,26].

When the dominant mechanism of electron-phonon coupling is of the density-displacement type, as in the local Holstein model [5], reaching large values of T_c is problematic [27–32]. The reason is the exponentially large effective bipolaron masses originating from small phonon overlap integrals for realistic values of the adiabatic parameter

$$\gamma = \frac{\omega_{\text{ph}}}{W} \ll 1,$$

where ω_{ph} is the phonon frequency and W is the particle bandwidth; $W \approx 4dt$ for the tight-binding dispersion relation with hopping amplitude t between the nearest-neighbor sites on a simple d -dimensional cubic lattice. The corresponding bipolarons are also very sensitive to the on-site Hubbard repulsion U . Much stronger electron-phonon interaction (EPI) is required for their formation when $U \sim W$, leading to an additional exponential increase in the effective mass [28,30–32]. These effects are less dramatic in the extended Holstein model with nonlocal EPI [6,33]. Nevertheless, Ref. [34] finds that in the adiabatic regime, the effective bipolaron masses remain too large to provide a viable high- T_c mechanism.

The prospects for obtaining high T_c appear to be far better when strong EPI originates from hopping dependence on atomic displacements by one of two mechanisms. In mechanism A, tunneling is enhanced (suppressed) when the distance between the sites is reduced (increased) [35,36]. In mechanism B, tunneling is modulated by displacements of atoms in the barrier region between the sites [37], as illustrated in Fig. 1. Previous studies [8,38,39] found that these bond polarons remain relatively light when entering the

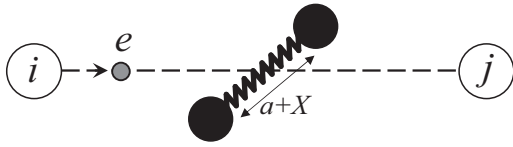


FIG. 1. An illustration of the coupling mechanism B when the electron hopping amplitude depends on the displacement of atoms in the vicinity of the tunneling path.

strong-coupling regime. However, properties of the corresponding bipolarons remain unexplored: The only available study [40] considered the $d = 1$ case in the antiadiabatic limit $\omega_{\text{ph}} = 3t$ for mechanism A. For realistic predictions of high T_c , one needs to look at higher-dimensional systems in the adiabatic limit $\gamma \ll 1$ and account for large repulsive potential between the electrons. This poses a significant computational challenge for unbiased methods based on exact diagonalization [28,33] or controlled truncation of the phonon Hilbert space [41] because bipolaron states in $d > 1$ are extended and the number of excited phonon modes quickly increases with $1/\gamma$. In this Research Letter, we show that path-integral representation for particles offers a unique opportunity for conducting comprehensive studies of singlet bipolaron states when EPI is based on mechanism B. The simplest Hamiltonian can be formulated as $\hat{H} = \hat{H}_e + \hat{H}_{\text{ph}} + \hat{H}_{\text{int}}$ with

$$\hat{H}_e = -t \sum_{\langle ij \rangle, \sigma} (\hat{c}_{j\sigma}^\dagger \hat{c}_{i\sigma} + \text{H.c.}) + \sum_{ij\sigma\sigma'} V_{\sigma\sigma'}(i, j) \hat{n}_{i\sigma} \hat{n}_{j\sigma'}, \quad (3)$$

$$\hat{H}_{\text{ph}} = \omega_{\text{ph}} \sum_{\langle ij \rangle} (\hat{b}_{\langle ij \rangle}^\dagger \hat{b}_{\langle ij \rangle} + 1/2), \quad (4)$$

$$\hat{H}_{\text{int}} = -g \sum_{\langle ij \rangle, \sigma} (\hat{c}_{j\sigma}^\dagger \hat{c}_{i\sigma} + \text{H.c.}) (\hat{b}_{\langle ij \rangle}^\dagger + \hat{b}_{\langle ij \rangle}), \quad (5)$$

where $V_{\sigma\sigma'}(i, j)$ is the potential of interelectron interaction and g is the strength of EPI. We use standard notations for creation and annihilation operators for electrons (on site i with spin σ) and optical phonons (on bonds $\langle ij \rangle$). The model can be further generalized to deal with several dispersive phonon modes and study effects of phonon dynamics and polarization. Here, we focus on describing the numerical method and present illustrative results for bipolaron states in a two-dimensional system. These results indicate that the search for high- T_c regimes requires comprehensive exploration of the model parameter space because compact states can easily end up to be heavy while light effective masses come at the price of larger pair radius; see Eq. (2). One counterintuitive effect is that the s -wave transition temperature may increase with the on-site repulsion U because less compact bipolarons are significantly lighter.

Basic relations. For a generic few-body system, the mathematical object containing all information about its ground-state properties and potentially allowing *sign-free* Monte Carlo simulations can be formulated as

$$O_{ba}(\tau) = \langle b | e^{-(\tau/2)\hat{H}} \hat{O} e^{-(\tau/2)\hat{H}} | a \rangle. \quad (6)$$

Here, $\hat{O}$ is a certain observable, $\hat{H}$ is the system's Hamiltonian, τ is an appropriately large imaginary-time interval, and $|a\rangle$ and $|b\rangle$ are any two states having finite overlap with the ground state $|g\rangle$. In the asymptotic limit of $\tau \rightarrow \infty$, the answer is dominated by the projection onto the ground

state when $O_{ba}(\tau)$ is given by the product of the ground-state expectation value of $\hat{O}$ and the universal (for all observables) propagator $\mathcal{G}_{ba}(\tau)$:

$$O_{ba}(\tau) \xrightarrow{\tau \rightarrow \infty} \langle g | \hat{O} | g \rangle \langle b | e^{-\tau\hat{H}} | a \rangle \equiv \bar{O}_g \mathcal{G}_{ba}(\tau). \quad (7)$$

If the states $|a\rangle$ and $|b\rangle$ are free of phonons, $\mathcal{G}_{ba}$ is the standard n -particle Green's function. Since $\mathcal{G}_{ba}(\tau) \equiv I_{ba}(\tau)$, where $\hat{I}$ is the identity operator, the expectation value of $\hat{O}$ in the ground state can be represented as

$$\bar{O}_g = \frac{O_{ba}(\tau)}{I_{ba}(\tau)} \equiv \frac{\sum_{ab} W_{ab} O_{ba}(\tau)}{\sum_{ab} W_{ab} I_{ba}(\tau)}. \quad (8)$$

This is a standard for Monte Carlo (MC) method setup when the stochastic sampling process is designed to sample the propagators $\mathcal{G}_{ba}$ while matrix elements of physical properties are taken care of by MC estimators. The extended version of the relation—with sums over any subsets of states $|a\rangle$ and $|b\rangle$ with arbitrary weights W_{ab} —adds flexibility and efficiency in designing updating schemes.

According to Eqs. (6) and (7), the imaginary-time dependence of $\mathcal{G}_{ba}$ also contains direct information about the ground-state energy E_g :

$$\mathcal{G}_{ba}(\tau) \xrightarrow{\tau \rightarrow \infty} \langle b | g \rangle \langle g | a \rangle e^{-\tau E_g}. \quad (9)$$

Moreover, by selecting state $|b\rangle$ or state $|a\rangle$, or both, to belong to a particular—non-ground-state—symmetry sector, we can employ (9) to determine the energy of the ground state in a given sector. In this way, $\mathcal{G}_{ba}(\tau)$ can be used to obtain the quasiparticle dispersion relation and, in particular, its effective mass.

A direct procedure of extracting m_* from $\mathcal{G}_{ba}$ is based on its universal long-time and large-distance behavior; see, e.g., Ref. [42]. If for each of the states $|b\rangle$ and $|a\rangle$ we introduce the “center-of-mass” positions $\mathbf{R}_b$ and $\mathbf{R}_a$, respectively, and define the relative displacement as $\mathbf{R} = \mathbf{R}_b - \mathbf{R}_a$, then

$$\mathcal{G}_{ba}(\tau \rightarrow \infty, \mathbf{R}) \rightarrow \frac{A_{ba} e^{-E_g \tau}}{\tau^{d/2}} e^{-\frac{m_* \mathbf{R}^2}{2\tau}}. \quad (10)$$

Apart from the coefficient A_{ba} , the right-hand side of (10) is insensitive to the particular choice of states $|a\rangle$ and $|b\rangle$, allowing one to average over them as in Eq. (8). Thus effective mass can be estimated from the mean-square fluctuations of the relative displacement at long τ :

$$\overline{\mathbf{R}^2}(\tau) = \frac{\sum_{ab} W_{ab} \mathcal{G}_{ba}(\tau, \mathbf{R}) \mathbf{R}^2}{\sum_{ab} W_{ab} \mathcal{G}_{ba}(\tau, \mathbf{R})} \xrightarrow{\tau \rightarrow \infty} \frac{d}{m_*} \tau. \quad (11)$$

Perturbative expansion. Our approach to MC sampling is based on the general scheme proposed in Ref. [43] and rendered here. Let $\hat{H}_0$ and $\hat{V}$ be the diagonal and off-diagonal parts of the Hamiltonian with respect to some basis $\mathcal{B} = \{|\alpha\rangle\}$ (our choice is site Fock states for particles and bond Fock states for phonons): $\hat{H}_0|\alpha\rangle = E_\alpha|\alpha\rangle$, $\langle\alpha|\hat{V}|\alpha\rangle = 0$. Next, we decompose $\hat{V}$ into elementary nonvanishing terms, $\hat{V} = \sum_{\alpha\beta} V_{\beta\alpha}|\beta\rangle\langle\alpha|$. In the chosen representation, there are three types of the elementary terms, each corresponding to the particle hopping along a specific bond in a specific direction: (i) bare hopping (with $V_{\beta\alpha} = -t$), (ii) hopping assisted by the phonon creation (with $V_{\beta\alpha} = -g\sqrt{n_\alpha + 1}$), and (iii) hopping

assisted by the phonon annihilation (with $V_{\beta\alpha} = -g\sqrt{n_\alpha}$). For a sign-free MC method—apart from, possibly, the negative signs (or phases) originating from components of the vectors $|a\rangle$ and $|b\rangle$ in the basis $\mathcal{B}$ —all matrix elements $-V_{\beta\alpha}$ need to be non-negative real numbers. This requirement is satisfied with our choice of $\mathcal{B}$ for the model given by (3)–(5).

Using a standard interaction representation for the evolution operator in the imaginary-time domain, $e^{-\tau\hat{H}} = e^{-\tau\hat{H}_0}\hat{\sigma}(\tau)$, and expanding $\hat{\sigma}(\tau)$ into a Taylor series, we arrive at [43]

$$\begin{aligned} \sigma_{\beta\alpha}(\tau) = & \delta_{\alpha\beta} - \int_0^\tau d\tau_1 V_{\beta\alpha} e^{\tau_1 E_{\beta\alpha}} \\ & + \sum_{\gamma_1} \int_0^\tau d\tau_2 \int_0^{\tau_2} d\tau_1 V_{\beta\gamma_1} e^{\tau_2 E_{\beta\gamma_1}} V_{\gamma_1\alpha} e^{\tau_1 E_{\gamma_1\alpha}} \\ & + \dots, \end{aligned} \quad (12)$$

where $E_{\beta\alpha} = E_\beta - E_\alpha$. The MC scheme is based on the statistical interpretation of the right-hand side of (12) viewed as an average over an ensemble of graphs representing strings of the hopping transitions, or “kinks,” $V_{\gamma_{i+1}\gamma_i}$. A string is characterized by the number and types of kinks, as well as by their space-time positions. Graphs are sampled according to their non-negative weights given by the values of the corresponding integrands in Eq. (12).

Sampling protocol and estimators. Our MC scheme includes τ in the configuration space of graphs and allows us, on the one hand, to sample the τ dependence of $\mathcal{G}_{ba}(\tau)$ and $\mathbf{R}^2(\tau)$ and then use Eqs. (9) and (13) to estimate the ground-state energy and the effective mass. On the other hand, this setup dramatically simplifies the minimalistic set of updates that deal exclusively with the last (in the τ domain) kink in the string. To be specific, we stochastically (i) add and remove the last bare-hopping kink using a pair of complementary updates [43], (ii) switch between the three types of hopping terms, and (iii) sample the length of the last time interval separating the last kink from the state $|b\rangle$. This scheme is ergodic and produces states $|b\rangle$ that admit any allowed configuration of excited phonon modes. Clearly, one can add additional updates dealing with other-than-last kinks for better decorrelation of the string configuration; see Ref. [43].

The proposed setup provides access to all the relevant system’s properties, including the same- and different-time correlation functions. They are measured when the value of τ is large enough and the asymptotic limit (7) is reached; its maximal value $\tau_{\max}$ is an important parameter controlling the accuracy of the projection onto the ground state. Monte Carlo estimators for observables are based on Eqs. (6) and (8) and their straightforward generalization for the different-time correlators. In accordance with the general theory (see, e.g., Ref. [43]), estimators do not involve additional computational costs (i.e., modifications of the configuration space and the sampling protocol) when the corresponding operators are either (i) diagonal in the basis $\mathcal{B}$ or (ii) expressed in terms of elementary $\hat{V}$ terms. The estimator for an observable of type (i) is simply the eigenvalue of this observable for the state $|\gamma\rangle \in \mathcal{B}$ created by the string of kinks at a certain moment of imaginary time, τ_* , close to $\tau/2$. The estimator for an observable of type (ii) is minus the total number of the

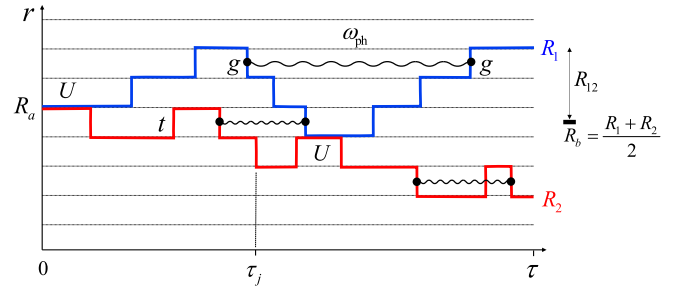


FIG. 2. Path-integral representation of the two-particle evolution with diagrammatic treatment of phonons.

corresponding $\hat{V}$ kinks within a certain time interval divided by the duration of this interval. It can be arbitrarily long provided its boundaries are appropriately far from the string end points $\tau = 0$ and $\tau \sim \tau_{\max}$.

Diagrammatics for the phonon sector. In the considered model, phonons do not interact with each other. This allows one to treat them diagrammatically, while keeping the path-integral representation for the particle sector only. The benefit of the diagrammatic representation for phonons (compared with their path-integral treatment described above) is the flexibility of dealing with dispersive modes. Here, the phonon dispersion is accounted for by simply modifying the zero-temperature (to comply with the diagrammatic rule that all averages be specified in terms of the vacuum state) phonon propagators, $D_s(\mathbf{r}_1, \tau_1; \mathbf{r}_2, \tau_2) \equiv D_s(\mathbf{r}_1 - \mathbf{r}_2, \tau_1, \tau_2)$, with $s = 1, \dots, d$ enumerating directions of lattice bonds. For sign-free formulation, D_s should be non-negative. One can still access (i) the particle configurations and (ii) the distribution of phonon numbers, as well as the correlations between items (i) and (ii). The path-integral treatment of dispersive phonons is also possible and comes at a price of dealing with yet another family of kinks originating from phonon hopping. The benefit of this approach is detailed information about spatial configurations of excited phonon modes.

The mathematical structure of the perturbative expansion with diagrammatic treatment of phonons is similar to the series (12) provided the Greek subscripts are used exclusively for the particle states. Accordingly, E_α now refer to energies of the particle subsystem alone, while each kink in (12) representing the phonon-assisted hopping event is associated with one of the two end points of the vacuum phonon propagator $D_s(\mathbf{r}_1, \tau_1; \mathbf{r}_2, \tau_2)$. The other end belongs either to states $|a\rangle$ or $|b\rangle$ or to another phonon-assisted hopping event along the bond with the same direction s . A graphical representation is shown in Fig. 2.

Illustrative results for singlet bipolarons. For estimates of T_c , apart from the bipolaron energy and effective mass, we determine R_*^2 from the probability distribution $P(R_{12})$ of finding particles at distance R_{12} from their center-of-mass position

$$R_*^2 = \sum_{R_{12}} R_{12}^2 P(R_{12}). \quad (13)$$

Figure 3 shows the dependence of the ground-state energy and the effective mass on the strength of the on-site repulsion for coupling constant $g = t/\sqrt{2}$ and the adiabatic parameter $\gamma = 1/16$. The ground-state energies are found to be smaller

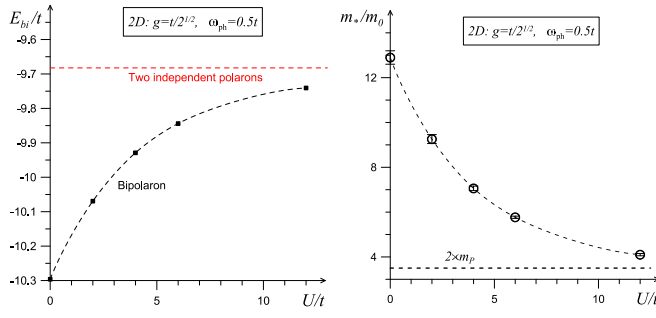


FIG. 3. Bipolaron energies (left) and effective masses (right) as functions of on-site repulsion U for coupling constant $g = t/\sqrt{2}$ and adiabatic parameter $\gamma = 1/16$. Error bars for E_{bi} are much smaller than symbol size. Dashed lines in all plots are used as guides to the eye.

than the energies of two polarons for any realistic value of U , indicating that we are dealing with bound states (bipolarons). At $U = 0$, the bipolaron effective mass is more than three times larger than its asymptotic limit $2m_p$, where m_p is the mass of a single polaron. With increasing U , the effective mass drops significantly, and at $U \gtrsim 10$ it approaches the limiting value of $2m_p$.

In contrast to naive expectations that a significant drop in the effective mass should result in a substantial increase in T_c , the trend revealed in Fig. 4 is quite different. The estimate for the critical temperature remains relatively flat up to $U \sim 2$ and then starts to decrease, dropping by almost a factor of 3 at $U \sim 10$. This is because the decrease in m_* is accompanied by a rapid increase in R_* , so that the product $m_* R_*^2$ does not show a pronounced maximum at any finite U .

A pronounced maximum of T_c at moderate values of U does not exist for large values of the coupling constant, as illustrated by the data for bipolaron states at $g = t$ and $\gamma = 1/16$ in Figs. 5 and 6. Here, the behavior of both the ground-state energy and the effective mass is qualitatively similar to what we had at $g = t/\sqrt{2}$ (see Fig. 3). However, the increase in R_* with U is not as dramatic and does not overcompensate the decrease in m_* even at $U \sim 20$. The maximum of T_c at $U > 24$ with the maximal value of T_c significantly larger than at $U = 0$ does exist because for $U \rightarrow \infty$ the energy of the delocalized bipolaron state is extremely close to the bound state threshold.

Conclusions. The model given by (3)–(5) of bond (bi)polarons (in any spatial dimension) allows a controlled and efficient numeric solution by Monte Carlo methods

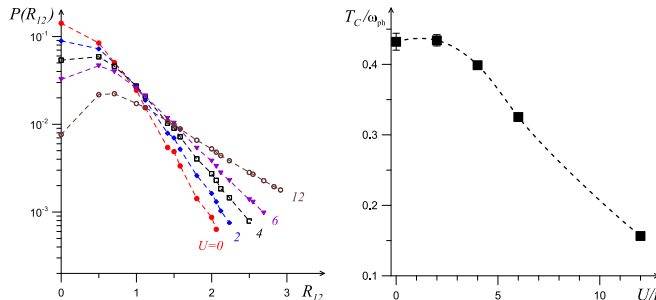


FIG. 4. Radial profiles for bipolaron states (left) and estimates of T_c (right) based on Eq. (2) for the same set of parameters as in Fig. 3.

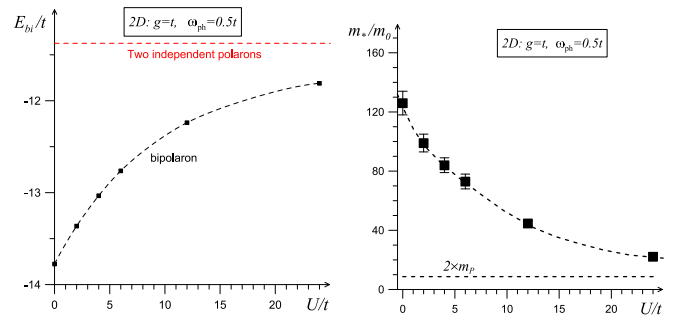


FIG. 5. Bipolaron energies (left) and effective masses (right) as functions of on-site repulsion U for coupling constant $g = t$ and adiabatic parameter $\gamma = 1/16$. Error bars for E_{bi} are much smaller than symbol size.

formulated in the path-integral representation for the electron(s) and either path-integral or real-space-diagrammatic representation for phonons. This is also true for the generalizations of (3)–(5) that include (i) dispersive bond phonons and (ii) additional density-displacement couplings as in the Holstein model and its extensions. For illustrative purposes, we presented simulations of singlet bipolaron states for the model given by (3)–(5) in two dimensions. Careful exploration of the multidimensional parameter space of the model, in search for the optimal T_c under realistic material conditions, is the subject of future work.

In the context of the bipolaron mechanism of high-temperature superconductivity, both the effective mass and the size of the bipolaron are equally important. Our results show that the positive effect of having a smaller effective mass can be readily overcompensated by the negative effect of having a larger bipolaron size; see Fig. 4.

There exists a range of parameters where the net effect of the strong repulsion between the electrons is a substantial increase in the critical temperature for the superconducting transition; see Fig. 6. One may find this result rather counter-intuitive given that normally the on-site repulsion suppresses Cooper pairing in the s channel.

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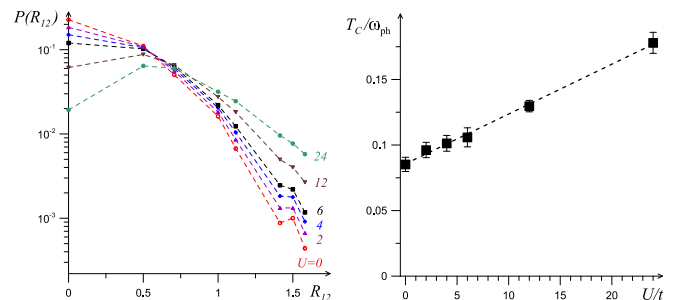


FIG. 6. Radial profiles for bipolaron states (left) and estimates of T_c (right) based on Eq. (2) for the same set of parameters as in Fig. 5.

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