Path Integral Description of a Semiclassical Su-Schrieffer-Heeger Model

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The electron motion along a chain is described by a continuum version of the Su-Schrieffer-Heeger Hamiltonian in which phonon fields and electronic coordinates are mapped onto the time scale. The path integral formalism allows us to derive the non local source action for the particle interacting with the oscillators bath. The method can be applied for any value of the e-ph coupling. The path integral dependence on the model parameters has been analysed by computing the partition function and some thermodynamical properties from $T = 1K$ up to room temperature. A peculiar upturn in the low temperature heat capacity over temperature ratio (pointing to a glassy like behavior) has been ascribed to the time dependent electronic hopping along the chain.

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1. INTRODUCTION

Conjugated organic polymers have been a focus of intensive research over the last twentyfive years after the discovery of their remarkable conducting properties tuned by the dopant concentration [1]. Undoped trans-polyacetylene sustains neutral solitons in the dimerized chain as a consequence of the Peierls instability [2]. Charge injection in polymers induces a lattice distortion with the associated formation of localized excitations, polarons and/or charged solitons. While the latter can exist only in trans-polyacetylene whose ground state is twofold degenerate, the former solutions are more general since they do not require such degeneracy. The Su-Schrieffer-Heeger (SSH) model Hamiltonian [3] is the fundamental tool in the analysis of quasi one dimensional polymers. In the weak coupling regime a continuum version [4] of the SSH model has been developed and polaronic solutions have been obtained analytically [5]. Although the 1D properties of the SSH model have been mainly investigated so far [6–11], extensions to two dimensions were considered in the late eighties in connection with the Fermi surface nesting effect on quasi 2D high $T_c$ superconductors [12]. Recent numerical analysis [13] have revealed the rich physics of 2D-SSH polarons whose mass seems to be larger than in the 1D case, at least in the intermediate regime of the adiabatic parameter [14].

One of the main issues in the study of e-ph models regards the possibility to monitor the conditions for polaron formation as a function of the coupling strength. The Feynman path integral method [15] is a landmark in this respect and several studies [16–18] have applied the path integral formalism both to the acoustical polaron of the Fröhlich Hamiltonian and to the Holstein model [19]. As a main property of the SSH model, the e-ph interaction modifies the bare electron hopping integral of the tight binding approximation thus leading to an Hamiltonian linearly dependent on the phonon displacement field. This feature will allow us to attack the SSH model.
by path integrals techniques after introducing a generalized version of the semiclassical model which treats only the electrons quantum mechanically. The general formalism is described in Section II together with the method to compute the full partition function of the interacting system. In Section III, we focus on the peculiarities of the free energy and heat capacity as a function of the model parameters. Section IV contains the final remarks.

II. PATH INTEGRAL FORMALISM

The 1D SSH interacting Hamiltonian reads:

\[
H = \sum_{r} J_{r,r+1} \left( f_{r}^\dagger f_{r+1} + f_{r+1}^\dagger f_{r} \right)
\]

\[
J_{r,r+1} = -\frac{1}{2} \left[ J + \alpha (u_{r} - u_{r+1}) \right]
\]  

(0.1)

where \( J \) is the nearest neighbors hopping integral for an undistorted chain, \( \alpha \) is the electron-phonon coupling, \( u_{r} \) is the dimerization coordinate which specifies the displacement of the monomer group on the \( r \)- lattice site along the molecular axis, \( f_{r}^\dagger \) and \( f_{r} \) create and destroy electrons (i.e., \( \pi \) band electrons in polyacetylene) on the \( r \)-group. Our non-interacting Hamiltonian is given by a set of independent oscillators which will be treated classically in the following. The real space Hamiltonian in eq.(1) can be transformed in a time dependent Hamiltonian by introducing \( x(\tau) \) and \( y(\tau') \) as the electron coordinates at the \( r \) and \( r+1 \) lattice sites, respectively. The spatial e-ph correlations contained in eq.(1) are then mapped onto the time axis by changing: \( u_{r} \rightarrow u(\tau) \) and \( u_{r+1} \rightarrow u(\tau') \). Accordingly, the time dependent Hamiltonian is:

\[
H(\tau, \tau') = J_{\tau,\tau'} \left( f_{\tau}^\dagger(x(\tau)) f_{\tau}(y(\tau')) + f_{\tau+1}^\dagger(y(\tau')) f_{\tau}(x(\tau)) \right)
\]

\[
J_{\tau,\tau'} = -\frac{1}{2} \left[ J + \alpha (u(\tau) - u(\tau')) \right]
\]  

(0.2)

The twofold degenerate ground state of the SSH Hamiltonian undergoes a Peierls instability which opens a gap at the Fermi energy. In real space the soliton connects the two degenerate phases with different senses of dimerizations and a localized electronic state is associated with each soliton. Both electron hopping between solitons and electron hopping to band states (thermal excitation) are in principle possible within the model. Mapping the Hamiltonian onto the time scale we maintain these properties while thermally activated electron hops become time dependent. \( H(\tau, \tau') \) is therefore more general than the real space SSH Hamiltonian since hopping processes are not constrained to first neighbors sites along the chain. We emphasize that a variable range hopping introduces some local disorder in the system.

Eq.(2) displays the semiclassical nature of the model in which quantum mechanical degrees of freedom interact with the classical variables \( u(\tau) \). Setting \( \tau' = 0 \), \( u(0) \equiv \)
where, $L$ is the chain length, $a$ is the lattice constant, $\beta$ is the inverse temperature, $\nu_n$ are the fermionic Matsubara frequencies and $\epsilon_k = -J \cos(k)$ is the electron dispersion relation. The chemical potential has been pinned to the zero energy level. After summing over the frequencies in eq.(3) we observe that the average energy per site can be written as

$$< H(\tau) > = \frac{1}{N} V(x(\tau)) + u(\tau) j(\tau)$$

with $N = L/a$ being the number of lattice sites and $n_F$ is the Fermi function. $V(x(\tau))$ is an effective term accounting for the $\tau$ dependent electronic hopping while $j(\tau)$ is the external source [20] current for the oscillator field $u(\tau)$. Averaging the electrons over the ground state we neglect the fermion-fermion correlations [21] which lead to effective polaron-polaron interactions in non perturbative analysis of the model [22]. This approximation however is not expected to affect substantially the following calculations.

Taking a large number of oscillators $(u_i(\tau), i = 1..N)$ as the bath for the quantum mechanical particle whose coordinate is $x(\tau)$, one can write the general path integral at any temperature as:

$$< x(\beta)|x(0) > = \prod_i \int Du_i(\tau) \int Dx(\tau) \cdot 
\exp \left[ -\frac{1}{\hbar} \int_0^{\hbar \beta} d\tau \sum_i \frac{M_i}{2} \left( \dot{u}_i^2(\tau) + \omega_i^2 u_i^2(\tau) \right) \right] \cdot 
\exp \left[ -\frac{1}{\hbar} \int_0^{\hbar \beta} d\tau \left( \frac{m}{2} \dot{x}^2(\tau) + V(x(\tau)) - \sum_i \gamma_i u_i(\tau) j(\tau) \right) \right]$$

where, $m$ is the electron mass, $M_i$ is the mass of the $i$-th ionic group, $\omega_i$ is the oscillator frequency, $\gamma_i$ is the coupling constant between external source term $j(\tau)$ and $i$-th oscillator $u_i$. In our model, $M_i \equiv M$ and $\gamma_i = 1$ hence, integrating out the oscillators coordinates over the
paths $Du_i(\tau)$, imposing a closure condition on the paths and replacing $\tau \to \tau/\hbar$, we get

$$< x(\beta)|x(0) > = \prod_i Z_i \int Dx(\tau) \cdot$$

$$\exp \left[ - \int_0^\beta d\tau \left( \frac{m}{2} x^2(\tau) + V(x(\tau)) \right) - \frac{1}{\hbar} A(x(\tau)) \right],$$

$$A(x(\tau)) = -\frac{\hbar^2}{4M} \sum_{i=1}^N \frac{1}{\hbar \omega_i \sinh(\hbar \omega_i \beta/2)} \cdot$$

$$\int_0^\beta d\tau \int_0^\beta d\tau' \cosh(\omega_i(|\tau - \tau'| - \beta/2)) j(\tau'),$$

$$Z_i = \frac{1}{2 \sinh(\hbar \omega_i \beta/2)} \quad \text{(0.6)}$$

The quadratic source term $A(x(\tau))$ is the non local (in time) action for the particle having memory of the interactions with the oscillators field. Assuming periodic conditions $x(\tau) = x(\tau + \beta)$, the particle paths can be expanded in Fourier components

$$x(\tau) = x_o + \sum_{n=1}^\infty 2 \left( \Re x_n \cos(\omega_n \tau) - \Im x_n \sin(\omega_n \tau) \right)$$

$$\omega_n = 2\pi n/\beta \quad \text{(0.7)}$$

and, taking the following measure of integration

$$\oint Dx(\tau) = \int_{-\infty}^{\infty} \frac{dx_o}{\sqrt{2\pi m K_B T}} \prod_{n=1}^\infty \left[ \frac{\int_{-\infty}^{\infty} d\Re x_n \int_{-\infty}^{\infty} d\Im x_n}{\pi \hbar^2 K_B T/m \omega_n^2} \right]$$

$$\quad \text{(0.8)}$$

a direct integration of eq.(6) can be carried out in order to derive the full partition function of the system versus temperature.

Let’s point out that, by mapping the electronic hopping motion onto the time scale, a continuum version of the interacting Hamiltonian (eq.(2)) has been de facto introduced. Unlike previous [23] approaches however, our path integral method is not constrained to the weak $e$-$ph$ coupling regime and it can be applied to any range of physical parameters.

II. COMPUTATIONAL METHOD AND RESULTS

As a preliminar step we determine, for a given path and at a given temperature: i) the minimum number ($N_k$) of $k$–points in the Brillouin zone to accurately estimate the average interacting energy per lattice site and, ii) the minimum number ($N_\tau$) of points in the double time integration to get a numerically stable source action in eq.(6). We find: $N_k = 70$ and $N_\tau = 300$ at $T = 1K$.

Computation of eqs.(6)-(8) requires fixing two sets of input parameters. The first set contains the physical
quantities characterizing the system: the bare hopping integral $J$, the oscillator frequencies $\omega_i$ and the effective coupling $\chi = \alpha^2 \hbar^2 / M$ (in units meV$^3$). The second set defines the paths for the particle motion which mainly contribute to the partition function through: the number of pairs $(\Re x_n, \Im x_n)$ in the Fourier expansion of eq.(7), the cutoff $(\Lambda)$ on the integration range of the expansion coefficients in eq.(8) and the related number of points $(N_\Lambda)$ in the measure of integration which ensures numerical convergence.

After introducing a dimensionless path

$$x(\tau)/a = \bar{x}_o + \sum_{n=1}^{N_p} \left( \bar{a}_n \cos(\omega_n \tau) + \bar{b}_n \sin(\omega_n \tau) \right), \quad (0.9)$$

with: $\bar{a}_n \equiv 2 \Re x_n / a$ and $\bar{b}_n \equiv -2 \Im x_n / a$, the functional measure of eq.(8) can be rewritten for computational purposes as:

$$\oint Dx(\tau) \approx \frac{a}{\sqrt{2}} \left( \frac{a}{2} \right)^{2N_p} \frac{(2\pi \cdot 4\pi \cdots 2N_p\pi)^2}{(\pi \hbar^2 / mK_B T)^{N_p+1/2}} \int_{-\Lambda/a}^{\Lambda/a} d\bar{x}_o \cdot 
\int_{-2\Lambda/a}^{2\Lambda/a} d\bar{a}_1 \int_{-2\Lambda/a}^{2\Lambda/a} d\bar{b}_1 \cdots \int_{-2\Lambda/a}^{2\Lambda/a} d\bar{a}_{N_p} \int_{-2\Lambda/a}^{2\Lambda/a} d\bar{b}_{N_p} \quad (0.10)$$

We take the lattice constant $a = 1 \text{Å}$. As a criterion to set the cutoff $\Lambda$ on the integration range, we notice that the functional measure normalizes the kinetic term in eq.(6):

$$\oint Dx(\tau) \exp \left[ -\frac{m}{2} \int_0^\beta d\tau \dot{x}^2(\tau) \right] \equiv 1, \quad (0.11)$$

and this condition holds for any number of pairs $N_p$ truncating the Fourier expansion in eq.(9). Then, taking $N_p = 1$ and using eq.(10), the left hand side transforms as:

$$\oint Dx(\tau) \exp \left[ -\frac{m}{2} \int_0^\beta d\tau \dot{x}^2(\tau) \right] \simeq \frac{4}{\pi} \left[ \int_0^U dy \exp(-y^2) \right]^2$$

$$U \equiv \sqrt{2\pi^3 \Lambda / \lambda}$$

$$\lambda = \sqrt{\frac{2\pi \hbar^2}{mK_B T}}. \quad (0.12)$$

Using the series representation [24]

$$\int_0^U dy \exp(-y^2) = \sum_{k=0}^{\infty} \frac{(-1)^k U^{2k+1}}{k!(2k+1)} = 0.886207$$

and taking $U = 3$ (the series converges with $k_{max} \sim 100$) eq.(13) yields 0.886207 which well approximates the Poisson integral value $\sqrt{\pi}/2$.

Thus, the cutoff $\Lambda$ can be expressed in terms of the thermal wavelength $\lambda$ as $\Lambda \sim 3\lambda / \sqrt{2\pi^3}$ hence, it scales
versus temperature as $\Lambda \propto 1/\sqrt{T}$. This means physically that, at low temperatures, $\Lambda$ is large since many paths are required to yield the correct normalization. For example, at $T = 1K$, we get $\Lambda \sim 284A$. Numerical investigation of eq.(6) shows however that a much shorter cutoff suffices to guarantee convergence in the path integral, while the cutoff temperature dependence implied by eq.(12) holds also in the computation of the interacting partition function. The thermodynamical results hereafter presented have been obtained by taking $\Lambda \sim \lambda/(10\sqrt{2\pi^3})$. Summing over $N_\Lambda \sim 20/\sqrt{T}$ points for each integration range and taking $N_p = 2$, we are then evaluating the contribution of $(N_\Lambda + 1)^{2N_p+1}$ paths (the integer part of $N_\Lambda$ is obviously selected at any temperature). Thus, at $T = 1K$ we are considering $\sim 4 \cdot 10^6$ different paths for the particle motion while, at $T = 100K$ the number of paths drops to 243. Low temperature calculations prove therefore to be extremely time consuming. Note that larger $N_p$ in the path Fourier expansion would further increase the computing time without introducing any substantial improvement in the thermodynamical output of our calculation.

Although the history of the SSH model is mainly related to wide band polymers, we take here a narrow band system ($J = 100meV$) to be consistent with previous investigations [11] and with the caveat that electron-electron correlations may become relevant in narrow bands. Free energy and heat capacity have been first (Figures 1-3) computed up to room temperature assuming a bath of $\bar{N} = 10$ low energy oscillators separated by $2meV$: $\bar{h}\omega_1 = 2meV, ..., \bar{h}\omega_{10} = 20meV$. The lowest energy oscillator yields the largest contribution to the phonon partition function mainly in the low temperature regime while the $\omega_{10}$ oscillator essentially sets the phonon energy scale which determines the size of the $e$-$ph$ coupling. A larger number $\bar{N}$ of oscillators in the aforegiven range would not significantly modify the calculation.

In the discrete SSH model, the value $\bar{\alpha} \equiv 4\alpha^2/(\pi\kappa J) \sim 1$, marks the crossover between weak and strong $e$-$ph$ coupling, with $\kappa$ being the effective spring constant. In our continuum and semiclassical model the effective coupling is the above defined $\chi$. Although in principle, discrete and continuum models may feature non coincident crossover parameters, we assume that the relation between $\alpha$ and $J$ obtained by the discrete model crossover condition still holds in our model. Hence, at the crossover we get: $\chi_c \sim \pi J h^2 \omega_{10}^2/64$. This means that, in Figures 1-3, the crossover value is set at $\chi_c \sim 2000meV^3$.

In Fig.1, the phonon free energy ($F_{ph}$) is plotted versus temperature together with the free energy due to the total action in eq.(6). $F_{sou}$ results from the competition between the free path action (kinetic term plus hopping potential in the exponential integrand) and the source action depending on the $e$-$ph$ coupling. While the former enhances the free energy the latter becomes dominant at increasing temperatures thus reducing the total free energy. As a main feature one notes that, by increasing
\( \chi \), \( F_{sou} \) gets a negative derivative at decreasing temperatures. In the very weak coupling case (\( \chi = 100 \text{meV}^3 \)) \( F_{sou} \) never intersects \( F_{ph} \) whereas an intersection point shows up both at moderately weak (\( \chi = 1000 \text{meV}^3 \)) and at moderately strong (\( \chi = 3000 \text{meV}^3 \)) couplings. The intersection temperature decreases as expected by increasing the strength of \( \chi \) but, at low \( T \) (< 50K), the contribution of the free electron path action prevails: this feature is reflected on the low \( T \) behavior of the heat capacity linear coefficient hereafter discussed.

Fig.2 shows the source term contributions to the heat capacity: the previously described summation over a large number of paths turns out to be essential to recover the correct thermodynamical behavior in the zero temperature limit. The small phonon contribution to the heat capacity is plotted in Fig.3 to point out that the Du-long Petit value is achieved at \( T \geq 200K \). Looking at the total heat capacity over temperature ratio, we find a peculiar low temperature upturn (also in the weak \( \chi \) regime) which can be mainly ascribed to the sizable effective hopping integral term \( V(x(\tau)) \). The e-ph coupling however determines the shape of the low \( T \) anomaly.

The oscillators bath affects the electronic correlations both on the space and time scales. Analysis of the thermal correlation function shows infact that high energy phonons substantially reduce the electronic correlation length at low temperatures while, in presence of low energy phonons, the electronic paths are correlated over the \( \tau \) scale. By increasing \( T \), the role of the phonons on the correlation function is less pronounced.

The effect of the oscillators bath on the thermodynamical properties is discussed in Figures 4-6 where the ten phonon energies are: \( \hbar \omega_1 = 22 \text{meV}, \ldots, \hbar \omega_{10} = 40 \text{meV} \). Accordingly the crossover is set at \( \chi_c \sim 8000 \text{meV}^3 \) and three plots out of five lie in the strong e-ph coupling regime. As shown in Fig.4, much larger \( \chi \) values (with respect to Fig.1) are required to get strongly decreasing free energies versus temperature while the \( \chi = 3000 \text{meV}^3 \) curve now hardly intersects the phonon free energy at room temperature. Fig.5 shows the rapid growth of the source heat capacity versus temperature at strong couplings whereas the presence of the low \( T \) upturn in the total heat capacity over \( T \) ratio is confirmed in Fig.6. Note that, due to the enhanced oscillators energies, the phonon heat capacity saturates at \( T \sim 400K \).

According to our integration method (eq.(10), at any temperature, a specific set of Fourier coefficients defines the ensamble of relevant particle paths over which the hopping potential \( V(x(\tau)) \) is evaluated. This ensamble is therefore \( T \) dependent. However, given a single set of path parameters one can monitor the \( V(x(\tau)) \) behavior versus \( T \). It turns out that the hopping decreases (as expected) by lowering \( T \) but its value remains appreciable also at low temperatures (< 20K). Since the \( d\tau \) integration range is larger at lower temperatures, the overall hopping potential contribution to the total action is relevant also at low \( T \). It is precisely this property which is responsible for the anomalous upturn in the heat capac-
ity linear coefficient. Further investigation also reveals that the upturn persists both in the extremely narrow ($J \sim 10meV$) and in the wide band ($J \sim 1eV$) regimes. Our computation method accounts for a variable range hopping on the $\tau$ scale which corresponds physically to introduce some degree of disorder along the linear chain. This feature makes our model more general than the standard SSH Hamiltonian (eq.(1)) with only real space nearest neighbor hops. While hopping type mechanisms have been suggested [25] to explain the striking conducting properties of doped polyacetylene at low temperatures we are not aware of any direct computation of specific heat in the SSH model. Since the latter quantity directly probes the density of states and integrating over $T$ the specific heat over $T$ ratio one can have access to the experimental entropy, our method may provide a new approach to analyse the transition to a disordered state which indeed exists in polymers [26]. In this connection it is also worth noting that the low $T$ upturn in the specific heat over $T$ ratio is a peculiar property of glasses [27,28] in which tunneling states for atoms (or group of atoms) provide a non magnetic internal degree of freedom in the potential structure [29].

FIG. 1. Phonon and Source Term contributions to the free energy for three values of the effective coupling $\chi$ (in units $meV^3$) and a narrow electron band. A bath of ten phonon oscillators is considered, the largest phonon energy being $\hbar \omega_{10} = 20meV$. 
FIG. 2. Source Term contributions to the heat capacity for the same parameters as in Fig.1.

FIG. 3. Total heat capacity over temperature for the same parameters as in Fig.1. The phonon heat capacity is also plotted.
FIG. 4. Phonon and Source Term contributions to the free energy for five values of the effective coupling $\chi$ (in units $meV^3$) and a narrow electron band. A bath of ten phonon oscillators has been taken, the largest phonon energy is $\hbar\omega_{10} = 40meV$.

FIG. 5. Source Term contributions to the heat capacity for the same parameters as in Fig.4.
FIG. 6. Total heat capacity over temperature for the same parameters as in Fig.4. The phonon heat capacity is also plotted.

III. CONCLUSIONS

Mapping the real space Su-Schrieffer-Heeger model onto the time space we have developed a semiclassical version of the interacting model Hamiltonian which can be attacked by path integrals methods. The acoustical phonons of the standard SSH model have been replaced by a set of oscillators providing a bath for the electron interacting with the displacements field. Time retarded interactions are naturally introduced in the formalism through the source action $A(x(\tau))$ which depends quadratically on the bare $e-ph$ coupling strength $\alpha$. Via calculation of the electronic motion path integral, the partition function can be derived in principle for any value of $\alpha$ thus avoiding those limitations on the $e-ph$ coupling range which burden any perturbative method. Particular attention has been paid to establish a reliable and general procedure which allows one to determine those input parameters intrinsic to the path integral formalism. It turns out that a large number of paths is required to carry out low temperature calculations which therefore become highly time consuming. The physical parameters have been specified to a narrow band system and the behavior of some thermodynamical properties, free energy and heat capacity, has been analysed for some values of the effective coupling strength lying both in the weak and in the strong coupling regime. We find a peculiar upturn in the low temperature plots of the heat capacity over temperature ratio indicating that a glassy like behavior can arise in the linear chain as a consequence of a time dependent electronic hopping with variable range.
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