Path Integral of the Two Dimensional Su-Schrieffer-Heeger Model

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The equilibrium thermodynamics of the two dimensional Su-Schrieffer-Heeger Model is derived by means of a path integral method which accounts for the variable range of the electronic hopping processes. While the lattice degrees of freedom are classical functions of time and are integrated out exactly, the electron particle paths are treated quantum mechanically. The free energy of the system and its temperature derivatives are computed by summing at any $T$ over the ensemble of relevant particle paths which mainly contribute to the total partition function. In the low $T$ regime, the heat capacity over $T$ ratio shows an upturn peculiar of a glassy like behavior. This feature is more sizeable in the square lattice than in the linear chain as the overall hopping potential contribution to the total action is larger in higher dimensionality.

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I. INTRODUCTION

The lattice dimensionality is a key parameter in materials science. One dimensional (1D) systems with half filled band undergo a structural distortion [1] which increases the elastic energy and opens a gap at the Fermi surface thus lowering the electronic energy. The competition between lattice and electronic subsystems stabilizes the 1D structure which accordingly acquires semiconducting properties whereas the behavior of the 3D system would be metallic like. Conjugated polymers, take polyacetylene as prototype, show anisotropic electrical and optical properties [2] due to intrinsic delocalization of $\pi$ electrons along the chain of CH units. As the intra-chain bonding between adjacent CH monomers is much stronger than the interchain coupling the lattice is quasi-1D. Hence, as a result of the Peierls instability, polyacetylene shows an alternation of short and long neighboring carbon bonds, a dimerization, accompanied by a two fold degenerate ground state energy. The Su-Schrieffer-Heeger (SSH) model Hamiltonian [3] has become a successful tool in polymer physics as it hosts the peculiar ground state excitations of the 1D conjugated structure and it accounts for a broad range of polymer properties [4]. As a fundamental feature of the SSH Hamiltonian the electronic hopping integral linearly depends on the relative displacement between adjacent atomic sites thus leading to a nonlocal $e$-$ph$ coupling with vertex function depending both on the electronic and the phononic wave vector. The latter property induces, in the Matsubara formalism [5], an electron hopping associated with a time dependent lattice displacement. As a consequence time retarded electron-phonon interactions arise in the system yielding a source current which depends both on time and on the electron path coordinates. This causes large $e$-$ph$ anharmonicities in the equilibrium thermodynamics of the SSH model [6]. Hopping of electrons from site
to site accompanied by a coupling to the lattice vibration modes is a fundamental process [7] determining the transport [8] and equilibrium properties [9] of many body systems. A variable range hopping may introduce some degree of disorder thus affecting the charge mobility [10] and the thermodynamic functions.

This paper focusses on this issue, dealing with the thermodynamical properties of the SSH model in two dimensions and comparing them with the results obtained in one dimension [11]. Only a few extensions of the SSH Hamiltonian to higher dimensionality appear in the literature [12] mainly concerning the phase diagrams [13] and the ground state excitations [14–16]. We apply a path integral method [17] which fully accounts for the time retarded e-ph interactions and, exploiting the above mentioned Hamiltonian linear dependence on the atomic displacement, allows us to derive the electron-phonon source action in two dimensions. The general formalism is outlined in Section II while the results are reported on in Section III. The conclusions are drawn in Section IV.

II. PATH INTEGRAL FORMALISM

In a square lattice with isotropic nearest neighbors hopping integral $J$, the SSH Hamiltonian for electrons plus e-ph interactions reads:

$$H = \sum_{r,s} \left[ (J_{r,s})_x (f_{r+1,s}^d f_{r,s} + h.c.) + (J_{r,s})_y (f_{r,s+1}^d f_{r,s} + h.c.) \right]$$

$$(J_{r,s})_x = -\frac{1}{2} [J - \alpha \Delta u_x]$$

$$(J_{r,s})_y = -\frac{1}{2} [J - \alpha \Delta u_y]$$

$$\Delta u_x = u_x(r + 1, s) - u_x(r, s)$$

$$\Delta u_y = u_y(r, s + 1) - u_y(r, s)$$

(0.1)

where $\alpha$ is the electron-phonon coupling, $\mathbf{u}(r, s)$ is the dimerization coordinate indicating the displacement of the monomer group on the $(r, s)$– lattice site, $f_{r,s}^d$ and $f_{r,s}$ create and destroy electrons (i.e., $\pi$ band electrons in polyacetylene). The phonon Hamiltonian is given by a set of 2D classical harmonic oscillators. The two addenda in (1) deal with one dimensional e-ph couplings along the $x$ and $y$ axis respectively, with first neighbors electron hopping. Second neighbors hopping processes (with overlap integral $J^{(2)}$) may be accounted for by adding to the Hamiltonian the term $H^{(2)}$ such that

$$H^{(2)} = (J_{r,s})_{x,y} (f_{r+1,s+1}^d f_{r,s} + h.c.)$$

$$(J_{r,s})_{x,y} = -\frac{1}{2} \left[ J^{(2)} - \alpha \sqrt{(\Delta u_x)^2 + (\Delta u_y)^2} \right]$$

(0.2)
The real space Hamiltonian in (1) can be transformed into a time dependent Hamiltonian [18] by introducing the electron coordinates: i) \( (x(\tau'), y(\tau')) \) at the \((r, s)\) lattice site, ii) \( (x(\tau), y(\tau')) \) at the \((r + 1, s)\) lattice site and iii) \( (x(\tau'), y(\tau)) \) at the \((r, s + 1)\) lattice site, respectively. \( \tau \) and \( \tau' \) vary on the scale of the inverse temperature \( \beta \).

The spatial e-ph correlations contained in (1) are mapped onto the time axis by changing:

\[
\begin{align*}
&u_{x,r,s}(\tau') \rightarrow u_{x,r,s}(\tau) \\
&u_{x,r+1,s} \rightarrow u_{x,r,s}(\tau) \\
&u_{y,r,s+1} \rightarrow u_{y,r,s}(\tau)
\end{align*}
\]

Now we set \( \tau' = 0 \), \( (x(\tau'), y(\tau')) \equiv (0, 0) \), \( (u_{x,\tau'}, u_{y,\tau'}) \equiv (0, 0) \).

Accordingly, (1) transforms into the time dependent Hamiltonian:

\[
H(\tau) = J_x(\tau) \left( \langle f^\dagger(x(\tau), 0) f(0, 0) + h.c. \rangle \right) + J_y(\tau) \left( \langle f^\dagger(0, y(\tau)) f(0, 0) + h.c. \rangle \right)
\]

\[
J_x(\tau) = -\frac{1}{2} \left[ J - \alpha u_x(\tau) \right]
\]

\[
J_y(\tau) = -\frac{1}{2} \left[ J - \alpha u_y(\tau) \right]
\]

(0.3)

While the ground state of the 1D SSH Hamiltonian is twofold degenerate, the degree of phase degeneracy is believed to be much higher in 2D [19] as many lattice distortion modes contribute to open the gap at the Fermi surface. Nonetheless, as in 1D, these phases are connected by localized and nonlinear excitations, the soliton solutions. Thus, also in 2D both electron hopping between solitons [20] and thermal excitation of electrons to band states may take place within the model. These features are accounted for by the time dependent version of the Hamiltonian.

As \( \tau \) varies continuously on the \( \beta \) scale and the \( \tau \)-dependent displacement fields are continuous variables (whose amplitudes are in principle unbound in the path integral), long range hopping processes are automatically included in \( H(\tau) \) which is therefore more general than the real space SSH Hamiltonian in (1) (and (2)). Thus by means of the path integral formalism we look at the low temperature thermodynamical behavior both in 1D and 2D searching for those features which may be ascribable to some local disorder related to the variable range of the hopping processes.

The semiclassical nature of the model is evident from (3) in which quantum mechanical degrees of freedom interact with the classical variables \( u_{x,y}(\tau) \). Averaging the electron operators over the ground state we obtain the time dependent semiclassical energy per lattice site \( N \):

\[
\frac{\langle H(\tau) \rangle}{N} = J_x(\tau) P(J, \tau, x(\tau)) + J_y(\tau) P(J, \tau, y(\tau))
\]

\[
P(J, \tau, v(\tau)) = \frac{1}{\pi^2} \int d\mathbf{k} \cos[\mathbf{k} \cdot v(\tau)] \cosh(\epsilon_k \tau) n_F(\epsilon_k)
\]

(0.4)

with \( v(\tau) = (x(\tau), 0) \) and \( v(\tau) = (0, y(\tau)) \) in the first and second addendum respectively.
\[ -J \sum_{i=x,y} \cos(k_i) \] is the electron dispersion relation and \( n_F \) is the Fermi function. Eq. (4) can be rewritten in a way suitable to the path integral approach by defining

\[
\langle H(\tau) \rangle_N = V(x(\tau)) + V(y(\tau)) + \mathbf{u}(\tau) \cdot \mathbf{j}(\tau)
\]

\[ V(x(\tau)) = -JP(J, \tau, x(\tau)) \]

\[ V(y(\tau)) = -JP(J, \tau, y(\tau)) \]

\[ \mathbf{j}(\tau) = \alpha \mathbf{P}(J, \tau, \mathbf{v}(\tau)) \]

\[ \mathbf{u}(\tau) = (u_x(\tau), u_y(\tau)) \]

(0.5)

\( V(x(\tau)) \) and \( V(y(\tau)) \) are the effective terms accounting for the \( \tau \) dependent electronic hopping while \( \mathbf{j}(\tau) \) is interpreted as the external source [21] current for the oscillator field \( \mathbf{u}(\tau) \). Averaging the electrons over the ground state we neglect the fermion-fermion correlations [22]. This approximation however is not expected to affect substantially the following calculations. Taking a bath of \( N \) 2D oscillators, we generally write the SSH electron path integral, \( \zeta(\tau) \equiv (x(\tau), y(\tau)) \), as:

\[
< \zeta(\beta) \mid \zeta(0) > \approx \prod_{i=1}^{N} \int D\mathbf{u}_i(\tau) \int D\zeta(\tau)
\]

\[
\cdot \exp \left[ -\int_{0}^{\beta} d\tau \sum_{i=1}^{N} \frac{M}{2} \left( \dot{u}_i^2(\tau) + \omega_i^2 u_i^2(\tau) \right) \right]
\]

\[
\cdot \exp \left[ -\int_{0}^{\beta} d\tau \left( \frac{m}{2} \dot{x}^2(\tau) + V(x(\tau)) + V(y(\tau)) \right) \right]
\]

\[
+ \sum_{i=1}^{N} \mathbf{u}_i(\tau) \cdot \mathbf{j}(\tau) \]

(0.6)

where \( m \) is the electron mass, \( M \) is the atomic mass and \( \omega_i \) is the oscillator frequency. As a main feature we notice that the interacting energy is linear in the atomic displacement field. Then, the electronic path integral can be derived after integrating out the oscillator degrees of freedom which are decoupled along the \( x \) and \( y \) axis. Thus, we get:

\[
< \zeta(\beta) \mid \zeta(0) > = \prod_{i=1}^{N} Z_i \left[ \int Dx(\tau) \right.
\]

\[
\cdot \exp \left[ -\int_{0}^{\beta} d\tau \left( \frac{m}{2} \dot{x}^2(\tau) + V(x(\tau)) \right) - \frac{1}{\hbar} A(x(\tau)) \right] \right]^2
\]

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

\[
\cdot \int_{0}^{\beta} d\tau j(x(\tau)) \int_{0}^{\beta} d\tau' \cosh \left( \hbar \omega_i (|\tau - \tau'| - \beta/2) \right) j(x(\tau'))
\]

\[
Z_i = \frac{1}{\left[ 2 \sinh(\hbar \omega_i \beta/2) \right]^2}
\]
and the 2D electron path integral is obtained after squaring the sum over one dimensional electron paths. This permits to reduce the computational problem which is nonetheless highly time consuming particularly in the low temperature limit. Note in fact that the source current \( j(x(\tau)) \) requires integration over the 2D Brillouin Zone (BZ) according to (4) and (5) and this occurs for any choice of the electron path coordinates. The quadratic (in the coupling \( \alpha \)) source action \( A(x(\tau)) \) is time retarded as the particle moving through the lattice drags the excitations of the oscillator fields which take a time to readjust to the electron motion. When the interaction is sufficiently strong the conditions for polaron formation [23] may be fulfilled in the system according to the degree of adiabaticity [24]. However the present path integral description is valid independently of the existence of polarons as it applies also to the weak coupling regime. Assuming periodic conditions \( x(\tau) = x(\tau + \beta) \), the particle paths can be expanded in Fourier components

\[
x(\tau) = x_o + \sum_{n=1}^{\infty} 2R \Re x_n \cos(\omega_n \tau) - 2\Im x_n \sin(\omega_n \tau)
\]

\[
\omega_n = 2\pi n/\beta
\]

and the open ends integral over the paths \( \int Dx(\tau) \) transforms into the measure of integration \( \oint Dx(\tau) \). Taking:

\[
\oint Dx(\tau) \equiv \int_{-\infty}^{\infty} \frac{dx_o}{(2\pi\hbar^2/mK_BT)^{1/2}} \prod_{n=1}^{\infty} \left[ \int_{-\infty}^{\infty} d\Re x_n \int_{-\infty}^{\infty} d\Im x_n \right] \]

which normalizes the kinetic energy term in (7), we proceed to integrate (7) in order to derive the full partition function of the system versus temperature.

III. THERMODYNAMICS IN 1D AND 2D

Taking a dimensionless path (in units of the lattice constant \( a = 1\AA \)) with: \( a_n \equiv 2R \Re x_n \) and \( b_n \equiv -2\Im x_n \) the functional measure in (9) can be rewritten as:

\[
\oint Dx(\tau) \approx \frac{2^{1/2}}{(2\lambda_m)^{2N_p}} \prod_{n=1}^{N_p} (2\pi n)^2 \int_{-\Lambda}^{\Lambda} dx_o \cdot \int_{-2\Lambda}^{2\Lambda} da_n \int_{-2\Lambda}^{2\Lambda} db_n
\]

\[
\lambda_m = \sqrt{\frac{\pi\hbar^2}{mK_BT}}
\]
path coefficients we first carry out the momentum integrations required by (4) summing over 1600 points in the reduced 2D BZ. This prepares the source current \( j(x(\tau)) \) to be used in the double time integration which yields, according to (7), the source action \( A(x(\tau)) \). Afterwards we sum the exponential of the total action over an appropriate set of particle paths. Numerically stable results are achieved by tuning the number of pairs \( N_p \) in (10), the cutoff \( \Lambda \) on the integration range of the Fourier coefficients in (10) and the related number of points \( (N_\Lambda) \) in the measure of integration. \( N_p = 2 \) suffices in the Fourier expansion of the path. The argument to set the cutoff \( \Lambda \) on the integration range is based on the fact that the functional measure normalizes the kinetic term in (7):

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

From (11) we get, \( \Lambda \sim 3\lambda_m/\sqrt{2\pi^3} \) hence, \( \Lambda \) scales versus temperature as \( \Lambda \propto 1/\sqrt{T} \). Computing (7) with (10) we find that in fact a smaller cutoff, \( \Lambda \sim \lambda_m/(10\sqrt{2\pi^3}) \), guarantees convergence in the interacting partition function. Accordingly the number of points in each integration range has to increase by decreasing \( T \) as \( N_\Lambda \propto 1/\sqrt{T} \). In the 2D problem, \( N_\Lambda \sim 35/\sqrt{T} \) points for each Fourier coefficient are required in (10). Then we carry out the total 2D path integral by evaluating the contribution of \( (N_\Lambda + 1)^2N_p^2 \) paths selecting the integer part of \( N_\Lambda \) at any temperature. Computation of the second order derivatives of the free energy in the range \( T \in [1, 301] K \), with a spacing of 3K, takes 55 hours and 15 minutes on a Pentium 4.

The physical input parameters characterizing both the 1D and 2D models are: the bare hopping integral \( J \), the oscillator frequencies \( \omega_i \) and the effective coupling \( \chi = a^2 \hbar^2/M \) (in units \( meV^3 \)). We take here a narrow band system with \( J = 100 meV \). The crossover between weak and strong e-ph coupling is given in our model by \( \chi_c \sim \pi \sqrt{\hbar^2 \omega_{max}^2/64} [2] \), where \( \omega_{max} \) is the largest energy in the oscillators bath. Let’s assume a bath of \( \bar{N} = 10 \) low energy oscillators with \( \hbar \omega_i \sim 20 meV \) thus setting the crossover value at \( \chi_c \sim 2000 meV^3 \). A higher number of oscillators in the same energy range would not significantly modify the results hereafter presented whereas lower \( \omega_i \) values would yield a larger contribution to the phonon partition function mainly at low \( T \). In Fig.1, a comparison between the 1D and the 2D free energies is presented for two values of \( \chi_c \), one lying in the weak and one in the strong e-ph coupling regime. The oscillator free energies \( F_{ph} \) are plotted separately while the free energies arising from the total action in (7), shortly termed \( F_{source} \), are due to the purely electronic plus e-ph contributions. In general, the 2D free energies have a larger gradient (versus temperature) than the corresponding 1D terms. The \( F_{ph} \) lie above the \( F_{source} \) both in 1D and 2D because of the choice of the \( \hbar \omega_i \); lowering the energy of
some oscillator modes would produce a crossing point between $F_{ph}$ and $F_{source}$ with temperature location depending on the value of $\chi$. Note that the $F_{source}$ plots have a positive temperature derivative in the low temperature regime and this feature is more pronounced in 2D. In fact, at low $T$, the source action is dominated by the hopping potential $V(x(\tau))$ while, at increasing $T$, the $e-ph$ effects become progressively more important (as the bifurcation between the $\chi_1$ and $\chi_2$ curves shows) and the $F_{source}$ get a negative derivative. In 2D, the weight of the $V(x(\tau))$ term is larger because there is a higher hopping probability. This physical expectation is taken into account by the path integral method. At any temperature, we monitor the ensemble of relevant particle paths over which the hopping potential is evaluated. For a selected set of Fourier components in (10) the hopping decreases by lowering $T$ but its value is still significant at low $T$. Considered that: i) the total action is obtained after a $d\tau$ integration of $V(x(\tau))$ and ii) the $d\tau$ integration range is larger at lower temperatures, we explain why the overall hopping potential contribution to the total action is responsible for the anomalous free energy behavior at low $T$.

In Fig.2, the heat capacity contributions due to the oscillators ($C_{ph}$) and electrons plus $e-ph$ coupling ($C_{sou}$) are reported on. The values are normalized over $\bar{N}$. The dimensionality effects are seen to be large and, for a given dimensionality, the role of the $e-ph$ interactions is magnified at increasing $T$. The total heat capacity ($C_{ph} + C_{sou}$) over $T$ ratios are plotted in Fig.3 in the low $T$ regime to emphasize the presence of an anomalous upturn which appears at $T \lesssim 10K$ in 1D and $T \lesssim 20K$ in 2D. This feature in the heat capacity linear coefficient is ultimately related to the above discussed overall hopping potential effects while the strength of the coupling $\chi$ has a minor role in the low $T$ limit.

Finally we emphasize that the degeneracy of the ground state scarcely affects the finite temperature equilibrium properties. This can be shown in 1D by treating the SSH Hamiltonian in mean field approximation with a staggered dimerization amplitude which yields the size of the lattice distortion. By diagonalizing the Hamiltonian in momentum space we see that the competition between electron and lattice energy determines the contribution to the partition function and shifts downwards the free energy versus temperature. Hence, the heat capacity due to the degeneracy is very small (in comparison with the values given in Fig.2) and strictly linear in $T$. Accordingly, the peculiar and sizeable upturn in the heat capacity over $T$ ratio does not depend on the nature of the ground state. The inclusion of the quantum lattice fluctuations beyond the mean field approximation is expected to reinforce our conclusion as the fluctuations tend to suppress the long range order associated with the dimerized structure [25].


IV. CONCLUSIONS

We have investigated the thermodynamics of a semi-classical Su-Schrieffer-Heeger (SSH) Hamiltonian extended to two dimensions in which the lattice displacements are taken as classical variables. By mapping the real space interactions onto the time ($\tau$) scale we deal with an Hamiltonian accounting for a variable range of the electronic transfer integral. Such a model can be suitably treated in the path integral formalism as both the electron and lattice degrees of freedom are continuous functions of $\tau$ which varies up to the inverse temperature. Although we are thus assuming a continuum model the present path integral calculation is valid for any range of e-ph coupling and for any value of physical parameters.

The two dimensional path integral for the SSH particle in a bath of harmonic oscillators has been derived by summing over a set of electron paths that becomes increasingly large at decreasing temperatures. Thus, the computation of the total action and of the free energy derivatives is highly time consuming in the low $T$ regime where the quantum mechanical nature of the electronic subsystem is fully accounted for. At low $T$, the electron hopping integral largely shapes the free energy which displays an anomalous positive derivative more pronounced in 2D than in 1D. This behavior is mirrored in the upturn of the heat capacity over $T$ ratio. For a given set of input parameters characterizing electron band, oscillator energies and e-ph couplings, we find that the upturn shows up at $T \lesssim 20K$ in the square lattice and at $T \lesssim 10K$ in the linear chain. Also the size of the upturn is larger in two dimensions consistently with the fact that the 2D hopping potential contribution to the total action is higher than in 1D. The presented results have been obtained assuming a narrow electron band ($J \sim 100meV$) but the upturn in the heat capacity over $T$ ratio appears also in the case of wide band systems.

The degenerate ground state of the SSH model is not responsible for the anomalous behavior of the heat capacity over $T$ ratio. In fact, closing the linear chain in a ring geometry, one can easily diagonalize the 1D Hamiltonian in mean field approximation and show that the contribution to the total heat capacity, due to the two fold degeneracy of the ground state, is small and linear versus temperature. A quantum description of the lattice displacement fields should further reduce the role of the dimerized ground state as the fluctuations tend to suppress the order parameter. Similar conclusions are expected to hold in the 2D case where, however, the degeneracy of the ground state is manyfold.

In conclusion, while in the low $T$ regime the thermodynamics is essentially governed by the electronic term of the Hamiltonian, at increasing $T$ the strength of e-ph coupling progressively determines the slope of the free energy and heat capacity. Our results suggest that a glassy like thermodynamical behavior is a peculiar feature of Hamiltonian models with a variable range for the
electronic hopping. Such a behavior is more likely to occur in higher dimensionality as the structure is more closely packed and, accordingly, the hopping probability is larger.

FIG. 1. Colour online. Phonon ($F_{ph}$) and Source Term ($F_{sou}$) contributions to the 1D and 2D free energies for two values of the effective coupling $\chi$: $\chi_1 = 1440\text{meV}^3$ (weak $e$-$\text{ph}$ coupling) $\chi_2 = 2560\text{meV}^3$ (strong $e$-$\text{ph}$ coupling).

FIG. 2. Colour online. Phonon and Source Term contributions (normalized over the number of oscillators) to the 1D and 2D heat capacities for the same parameters as in Fig.1. The oscillator heat capacities are also plotted.
FIG. 3. Colour online. Total heat capacity over temperature for the same parameters as in Fig.1.

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TOTAL HEAT CAPACITY / T (meV K^{-2})

TEMPERATURE (K)

- 2D $\chi_2$
- 2D $\chi_1$
- 1D $\chi_2$
- 1D $\chi_1$