Induced charge in a Fröhlich polaron: Sum rule and spatial extent

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Within the path-integral formalism, we derive exact expressions for correlation functions measuring the lattice charge induced by an electron and associated polarization in the Fröhlich polaron problem. We prove that a sum rule for the total induced charge, already obtained with approximate approaches, is indeed exact. As a consequence the total induced charge is shown rigorously to be temperature independent. In addition, we perform path integral Monte Carlo calculations of the correlation functions and compare them to variational results based on the Feynman method. As the temperature increases the polaron radius decreases. On the other hand, at high temperatures the electron motion is not hindered by the lattice. These apparently contradictory results are discussed.

I. INTRODUCTION

An electron added to an insulating polar crystal forms a quasiparticle called dielectric polaron after Fröhlich.[1,2] This has been recognized as a fundamental field theoretical problem.[3] More recently a variety of materials have emerged that present interesting properties when doped away from an insulating phase, like colossal magnetoresistent manganites and high-temperature superconducting cuprates. The fact that these are polar crystals has produced a renewed interest in Fröhlich polaron problems.[4–7]

Roughly speaking a dielectric polaron is composed of an electron and the (opposite) charge that it induces in the lattice. The electron and induced charge attract each other so that for the electron to move it has to drag the induced charge resulting in an increase of the quasiparticle mass. In this paper we study correlation functions that measure the magnitude and spatial extent of the induced charge and associated polarization field.

We derive a rigorous sum rule that states that the total induced charge equals the charge induced by a classical (static) electron and it is independent of temperature. This result, well known within perturbative[8] and variational approaches,[9] is proven here to be exact. The large distance behavior of the electric field is determined by this sum rule. In addition we discuss the short distance and the high-temperature asymptotic limit of these quantities. These results provide constraints to approximations on the polaron problem.

The real-space path-integral Monte Carlo (PIMC) method is used to evaluate correlation functions. These are compared with Feynman's variational approximation (FVA)[10,9] and analytical results in weak and strong coupling. We find that the polaron radius is determined at low temperatures by the electron-phonon coupling α alone while at high temperatures it is proportional to the de Broglie thermal wavelength ($\lambda_T = \hbar\sqrt{2\pi\beta/m}$ with β the inverse temperature) and becomes independent of coupling. We define a polaron crossover temperature $T^*(\alpha)$. Although the electron induces a temperature independent charge in the lattice the induced charge hinders the electron motion only below $T^*(\alpha)$. At high temperatures thermal effects wash out the hindering effect of the induced charge but, we remark again, not the induced charge itself.

II. ANALYTICAL RESULTS

Our starting point is the effective action for the Fröhlich polaron problem, after the phonons have been eliminated with the usual path integral techniques,[3] $S = S_0 + S_I$ with

$$S_0[\mathbf{x}] = \int_0^{\hbar\beta} d\tau \frac{1}{2} m\dot{x}^2, \tag{1}$$

$$S_I[\mathbf{x}] = -\frac{\alpha}{2\sqrt{2}} \frac{(\hbar\omega_L)^{3/2}}{m^{1/2}} \int_0^{\hbar\beta} d\tau_1 \int_0^{\hbar\beta} d\tau_2 \frac{D(\tau_1-\tau_2)}{|\mathbf{x}(\tau_1)-\mathbf{x}(\tau_2)|},$$

here m is the electron mass, $\alpha = e^2 m^{1/2}/\hbar\bar{\epsilon}(2\hbar\omega_L)^{1/2}$ is the coupling constant with ω_L the phonon frequency, $1/\bar{\epsilon} = 1/\epsilon_\infty - 1/\epsilon_0$ and ϵ_∞ (ϵ_0) is the high-(zero-) frequency dielectric constant.

$$D(\tau) = \frac{\exp(\omega_L|\tau|) + \exp[(\hbar\beta - |\tau|)\omega_L]}{\exp(\hbar\beta\omega_L) - 1} \tag{2}$$

is the phonon propagator.

We are interested in the correlation function between the electron charge density $-e\tilde{n}(\mathbf{r})$ and the charge induced in the lattice $e\tilde{n}_i(\mathbf{r})$ normalized to the probability density to find an electron at a given point, i.e., the inverse of the volume V. Dropping the charges this is defined as,

$$\langle \tilde{n}(0)\tilde{n}_i(\mathbf{r})\rangle / V^{-1} \equiv g(r)/\bar{\epsilon} \tag{3}$$

Averages are defined as path integrals weighted by S

$$\langle \ldots \rangle = \frac{\int \mathcal{D}\mathbf{x} e^{-S/\hbar}(\ldots)}{\int \mathcal{D}\mathbf{x} e^{-S/\hbar}} \tag{4}$$

where the paths entering in Eq. (4) depart and arrive at the same point. Further integration over such a point is not per-

formed and this assigns to Eq. (4) the meaning of a constrained average with $\mathbf{x}(0)=\mathbf{x}(\beta)=0$. Those averages are however equivalent to the unconstrained ones because of translational invariance. We used the spherical symmetry of the problem to define $g(r)$, and we divided by $\bar{\epsilon}$ on the right-hand side of Eq. (3) for later convenience. Another quantity of interest is the induced lattice polarization

$$\mathbf{P}(\mathbf{r})\equiv\langle\tilde{n}(0)\tilde{\mathbf{P}}(\mathbf{r})\rangle/V^{-1} \tag{5}$$

$\nabla\tilde{\mathbf{P}}(\mathbf{r})=-e\tilde{n}_i(\mathbf{r})$ is the density of polarization operator. The correlation function in Eq. (5) is related to the induced electrostatic potential $[\nabla^2 V(\mathbf{r})=-4\pi e g(r)/\bar{\epsilon}]$ considered in Ref. 11, which will not be discussed here. We stress that these quantities have the meaning of *correlation functions* measuring average induced charge, polarization, and induced potential at a distance r from the electron position.

The charge-density operator for the phonons is,[12]

$$e\tilde{n}_i(\mathbf{r})=-\sqrt{\frac{\hbar\omega_L}{4\pi V\bar{\epsilon}}}\sum_{\mathbf{k}} k\tilde{Q}_{\mathbf{k}}e^{i\mathbf{k}.\mathbf{r}}. \tag{6}$$

where $\tilde{Q}_{\mathbf{k}}$ is the dimensionless displacement for momentum $\mathbf{k}$ phonons. Inserting Eq. (6) in Eq. (3) we obtain an equation for $g(r)$ as a function of the density displacement correlation function $\langle\tilde{n}(0)\tilde{Q}_{\mathbf{k}}\rangle$. The phonon variables can be traced out by standard methods. We have found that it is possible to give an *exact* expression for the correlation functions in terms of path integrals weighted by the effective electronic action of Eq. (1). We find for the density-induced-density correlation function

$$g(r)=\int_0^{\hbar\beta} d\tau U(\tau)\langle\delta[\mathbf{r}-\mathbf{x}(\tau)]\rangle \tag{7}$$

and for the polarization field

$$\mathbf{P}(\mathbf{r})=-\frac{e}{4\pi\bar{\epsilon}}\int_0^{\hbar\beta} d\tau U(\tau)\left\langle\frac{\hat{\mathbf{r}}}{|\mathbf{r}-\mathbf{x}(\tau)|^2}\right\rangle, \tag{8}$$

where $\hat{\mathbf{r}}\equiv\mathbf{r}/r$ and

$$U(\tau)=\hbar\omega_L\frac{\sinh(\omega_L\tau)+\sinh[\omega_L(\hbar\beta-\tau)]}{2\tanh(\beta\hbar\omega_L/2)\sinh(\beta\hbar\omega_L)}. \tag{9}$$

Within FVA, the variational quadratic action can be exploited in Eqs. (7) and (8) to analytically perform the averages and to recover the results of Refs. 10 and 11.

Equations (7) and (8) have a simple physical interpretation. The induced charge can be seen as ''distributed'' along the electron path with weight $U(\tau)$. The polarization is the superposition of polarizations associated with those elementary induced charges.

Equation (7) can be integrated in the whole space using the properties of the Dirac's δ function. Since $\int d\tau U(\tau)=1$, we conclude that $g(r)$ is normalized to one. The total induced charge q is computed by integrating the density-induced density correlation function in Eq. (3):

$$q=e\int_0^{\infty} dr 4\pi r^2 g(r)/\bar{\epsilon}=\frac{e}{\bar{\epsilon}}, \tag{10}$$

which completes the proof of the sum rule. The total induced charge amounts to the charge the electron would induce if it where a static classical particle. In other words there are no quantum corrections to the total induced charge.

An alternative derivation can be worked out following Quémerais.[13] From the time derivative $i\hbar\ddot{\tilde{\mathbf{P}}}=[\dot{\tilde{\mathbf{P}}},\tilde{H}]$ one obtains

$$\ddot{\tilde{\mathbf{P}}}=\omega_L^2\left(\frac{1}{4\pi\bar{\epsilon}}\tilde{\mathbf{D}}-\tilde{\mathbf{P}}\right) \tag{11}$$

Here $\tilde{H}$ is the Hamiltonian, $\tilde{\mathbf{D}}$ is the electric displacement operator due to the electron ($\nabla\cdot\tilde{\mathbf{D}}=-4\pi e\tilde{n}$), and $i\hbar\dot{\tilde{\mathbf{P}}}=[\tilde{\mathbf{P}},\tilde{H}]$. Taking the divergence we obtain a relation for the charge operators:

$$\nabla\cdot\ddot{\tilde{\mathbf{P}}}(\mathbf{r})=\omega_L^2 e\left(-\frac{\tilde{n}(\mathbf{r})}{\bar{\epsilon}}+\tilde{n}_i(\mathbf{r})\right) \tag{12}$$

We can integrate this expression in all space and take the thermodynamic average. The left-hand side is proportional to the average of the net force felt by the lattice at the boundary of the system, which should vanish at equilibrium. The right-hand side gives Eq. (10).

Equation (10) shows that the induced charge is independent of temperature. This contradicts the naive argument that all polaron effects should disappear at high temperatures. To understand this behavior one can do an analogy with the behavior of an harmonic oscillator in an external field. In that case, because of harmonicity, one gets a displacement which is temperature independent. Here roughly speaking the harmonic oscillator represents the phonon coordinates and the external field is the field produced by the electron on the phonons. The induced charge is a measure of how much the ions displace from their bare equilibrium positions in the presence of the electron. As for the single harmonic oscillator, this ''displacement'' is independent of temperature. Only anharmonicities can make the induced charge temperature dependent.

Using the sum rule it is easy to see that at distances much larger than the polaron radius, as defined below, the polarization field goes as $\mathbf{P}(\mathbf{r})=-e\hat{\mathbf{r}}/(4\pi\bar{\epsilon}r^2)$. Clearly the distortion produced by the electron is long range, a fact that is not always recognized in the literature.[14] The total electric field (always in the sense of a correlation function) is given by $\mathbf{E}=\mathbf{D}-4\pi(\mathbf{P}|\mathbf{P}_\infty)$, where the electric displacement is given by $\mathbf{D}=-e\hat{\mathbf{r}}/r^2$ and the high-frequency polarization is $\mathbf{P}_\infty=-e(1-1/\epsilon_\infty)\hat{\mathbf{r}}/(4\pi r^2)$. At long distances we have $\mathbf{E}(\mathbf{r})=-e\hat{\mathbf{r}}/(\epsilon_0 r^2)$ which means that the electric field generated by the electron gets screened by the static dielectric constant. This is generally expected, but to the best of our knowledge, has never been proven for all coupling and temperatures.

Now we discuss the short-distance behavior. At distances much smaller than the polaron radius, we expect that the effect of the interaction becomes irrelevant in the functional integrals. This is because the latter are dominated by electron paths with short wavelength or equivalently high-kinetic en-

ergy. We can then replace the total action by the free-electron action in Eqs. (7) and (8). We obtain the asymptotic result

$$\lim_{r\to 0} \mathbf{P}(\mathbf{r}) = -\frac{e}{8\pi\epsilon \bar{l}^2 \tanh(\beta\hbar\omega_L/2)}\hat{\mathbf{r}}. \qquad (13)$$

where $l=\sqrt{\hbar/2m\omega_L}$ is the harmonic-oscillator characteristic length. Using the same argument we obtain that $g(r)\propto r^{-1}$ for $r\to 0$ and the proportionality coefficient can also be obtained with the same method. The latter behavior has been obtained in Ref. 9 within the FVA. These results coincide with lowest-order perturbation theory.

At high temperatures we also expect that the effect of the interaction becomes irrelevant and so we can replace again the total action by the free-electron action in the functional integrals. The high-temperature asymptotic result for $g(r)$ is

$$4\pi r^2 g(r) = \frac{2r}{l^2\beta\hbar\omega_L}\exp\left(\frac{-r^2}{l^2\beta\hbar\omega_L}\right) \beta\hbar\omega_L \ll 1. \qquad (14)$$

This result has also been obtained in Ref. 9 within FVA. We remark that although the density-induced-density correlation function does not vanish for large temperatures the polaron effective mass tends to the bare electron mass.

III. NUMERICAL RESULTS

Now we discuss the spatial extent of the induced charge at general couplings and temperatures. We have evaluated averages in Eq. (7) using PIMC. Equations (7) and (8), being expressed in real space rather than in Fourier components are more suitable for this purpose. We have performed Metropolis PIMC calculations within the imaginary time discretization scheme. In order to regularize the attractive divergence of the retarded action at short distance, and to improve the convergence with the number of the imaginary time discretization points, we have developed a preaveraging procedure similar to the one used for local actions.[15,16] Details of the method and more extensive results will be given in a separate publication.[17] Here we just say that the results for the $g(r)$ are well converged as checked by doubling the number of imaginary time slices. Previous MC studies of the Fröhlich polaron were limited to small ($\alpha\leqslant 4$) (Ref. 18) or intermediate ($\alpha\leqslant 7$) (Ref. 19) couplings and were focused to the calculation of the ground-state energy[18,19] and effective mass.[19]

Following Ref. 9 we have also computed $g(r)$ in the FVA, i.e., using Feynman's quadratic action to evaluate the average appearing in Eq. (7).[20]

In Fig. 1 we show $4\pi r^2 g(r)$ for weak, intermediate, and strong coupling. We also show the weak-coupling result obtained by perturbation theory at $T=0$ and the strong-coupling result in the Landau-Pekar approximation. For all couplings the correlation function decays exponentially with distance as expected for a polaron. The area under the curves is one according to the sum rule [Eq. (10)]. The short-distance asymptotic behavior Eq. (13) is exactly satisfied in FVA and is also satisfied in the PIMC within the numerical error. We define the polaron radius r_m as the distance at which $4\pi r^2 g(r)$ is a maximum. As the coupling increases

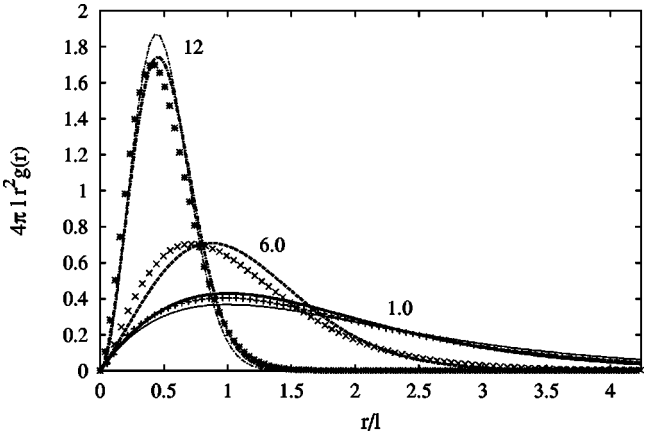

FIG. 1. $4\pi r^2 g(r)$ as a function of r for different couplings and approximations. From left to right bold curves (FVA after the formalism of Ref. 9) and points (PIMC) corresponds to $\alpha=12,6,1$, respectively, and $\beta\hbar\omega_L=20$. Thin-solid line is perturbation theory, thin-dashed line the Landau-Pekar strong-coupling approximation both at $\beta\hbar\omega_L=\infty$.

the polaron shrinks indicating the progressively more localized nature. In Fig. 2 we show the temperature dependence of $4\pi r^2 g(r)$ at intermediate coupling. In Figs. 1 and 2 we see that, apart from a small overestimate of the polaron radius, FVA reproduces fairly well the PIMC data. We can then safely use the FVA to discuss the spatial properties of the polaron. Notice that the good agreement found between PIMC and FVA is not obvious since in principle only the free energy is expected to be accurate for the latter.

Contrary to the naive expectation, the effect of temperature is to shrink the polaron (Fig. 2). Our physical explanation is the following: At low temperatures, phonons relax, shrinking the spatial extent of the electron until the increase in electron kinetic energy balances the gain in electron-phonon interaction energy. At high temperatures, however, a typical electron has energy $\bar{E}=3/2\beta$ and momentum $\hbar\bar{k}=\sqrt{3m/\beta}$. One can construct a wave packet of width Δk in momentum space using plane waves with higher and smaller

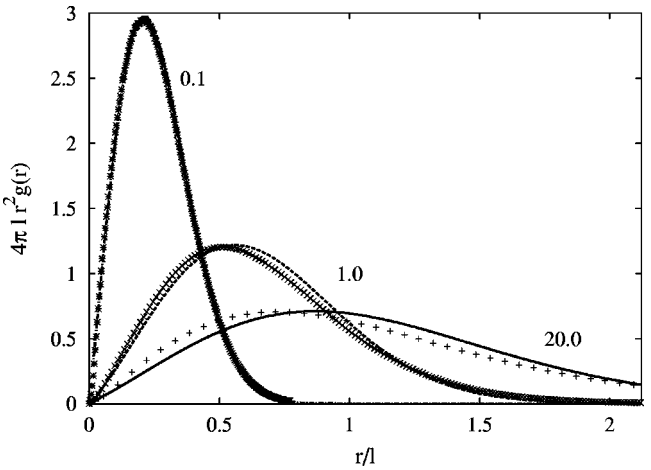

FIG. 2. $4\pi r^2 g(r)$ as a function of r at $\alpha=6$. From left to right curves (FVA after the formalism of Ref. 9) and points (PIMC) refers to $\beta\hbar\omega_L=0.1,1.0,20$, respectively.

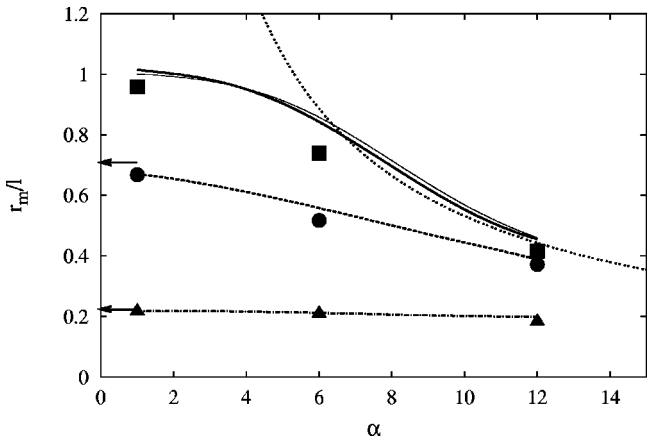

FIG. 3. The polaron radius as a function of coupling for different inverse temperatures. Results from FVA are shown as dash-dotted ($\beta\hbar\omega_L=0.1$), dashed ($\beta\hbar\omega_L=1.0$), thick-solid ($\beta\hbar\omega_L=20$), and thin-solid lines ($\beta\hbar\omega_L=\infty$). Dotted line is the Landau-Pekar approximation. Results from PIMC calculations are shown as triangles ($\beta\hbar\omega_L=0.1$), circles ($\beta\hbar\omega_L=1.0$), and squares ($\beta\hbar\omega_L=20$). The horizontal arrows indicate the value $0.2\lambda_T/l$ for $\beta\hbar\omega_L=1$ (upper) and $\beta\hbar\omega_L=0.1$ (lower). The arrows for $\beta\hbar\omega_L=20,\infty$ are out of scale.

energy without affecting the electron internal energy. The biggest Δk, which will not affect the electron internal energy, is of order of $\overline{k}$ itself. One can then achieve a localization of the electron of order $1/\overline{k}\sim\hbar/\sqrt{3m/\beta}\sim\lambda_T$. It follows that the phonons can relax at practically no cost until the polaron radius stabilizes at a value of this order. In fact at high temperatures, the asymptotic value of the polaron radius can be obtained from Eq. (14): $\lim_{\beta\to 0} r_m = l\sqrt{\beta\hbar\omega_L}/2 = 0.2\lambda_T$. This scaling has been found by Sethia *et al.*[21] for the mean-square displacement of the electron in imaginary time. Notice that the polaron radius becomes independent of the coupling.

To characterize the temperature and coupling dependence of the polaron size we plot in Fig. 3 the polaron radius as a function of coupling for different inverse temperatures. We see that when the temperature is such that $0.2\lambda_T > l$ (low temperatures, see the curves for $\beta\hbar\omega_L=20,\infty$), the polaron radius exhibits little temperature dependence at all couplings and simply interpolates between the weak-coupling polaron radius $r_m=l$ and the Landau-Pekar polaron radius $r_m = 3l\sqrt{\pi/2}/\alpha$. When $0.2\lambda_T<l$, two different regimes occur. At small coupling the polaron radius tends to saturate at $0.2\lambda_T$ (horizontal arrow) whereas at high couplings one recovers the low-temperature polaron radius $r_m(\alpha,T=0)$. We can define a crossover line when these two lengths are of equal magnitude so that the crossover temperature as a function of α is given by the equation $r_m(\alpha,T=0)=0.2\lambda_T(T^*)$. In the FVA approximation[17] we find the crossover temperature in energy units to be $T^*(\alpha)=0.15\hbar\omega_L v(\alpha,T=0)$ with v the usual Feynman variational parameter.[3] For $\alpha\to 0$, T^* goes to a constant of order of $0.5\hbar\omega_L$, whereas for large α it increases quadratically with α.

In the low-temperature regime ($T<T^*$) the polaron radius becomes almost temperature independent and is determined by the coupling.[22] The high-temperature regime $T>T^*$ is characterized by a polaron radius that is independent of the coupling and is determined by the temperature alone ($r_m=0.2\lambda_T$).

IV. CONCLUSIONS

We have studied the charge induced in the lattice by an electron in Fröhlich polaron problem and the associated polarization field. We have derived relations that express the charge-induced density correlation function in terms of path integral involving only the electronic degrees of freedom that are suitable to be evaluated by PIMC method. A rigorous sum rule was derived that determines the total induced charge and the long-distance behavior of the polarization field. We give also the asymptotic limits of these quantities at short distances and at high temperatures. We have compared results obtained using FVA through the lines of Refs. 9–11 and those obtained by PIMC method. To the best of our knowledge, this is the first PIMC computation of real-space correlation functions in a Fröhlich model. From the spatial dependence of the induced charge we obtained a polaron radius. The polaron radius is determined by the coupling at low temperatures and by the thermal wavelength at high temperatures with a crossover temperature that we evaluated in FVA.

At high temperature, a polaron with small radius and small effective mass is achieved. These results are not in contradiction because the small radius at high temperatures is a thermal effect of the electron and it is not related to the lattice response. The lattice acts only as a probe of the intrinsic electron localization radius, namely, λ_T. Obviously this small radius polaron has nothing to do with the Holstein zero-temperature small polaron that induces almost local lattice displacements and moves coherently with a large effective mass.

The PIMC polaron radius is always smaller than the FVA calculation in the range of coupling and temperature studied. This effect is more pronounced at intermediate couplings. The overall temperature dependence agrees with the findings of Refs. 9–11, however, our physical interpretation is different.

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[22] We remark that within FVA and for weak coupling, one finds a small increase of r_m with temperature with a shallow maximum (close to $\beta^{-1} \simeq 0.15 \hbar \omega_L$ for $\alpha = 1$) followed by a decrease at high temperatures according to the behavior discussed in the text.