Path-integral approach to resistivity anomalies in anharmonic systems

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Different classes of physical systems with sizeable electron-phonon coupling and lattice distortions present anomalous resistivity behaviors versus temperature. We study a molecular lattice Hamiltonian in which polaronic charge carriers interact with nonlinear potentials provided by local atomic fluctuations between two equilibrium sites. A path-integral model is developed to select the class of atomic oscillations which mainly contributes to the partition function, and the electrical resistivity is computed in a number of representative cases. We argue that the common origin of the observed resistivity anomalies lies in the time retarded nature of the polaronic interactions in the local structural instabilities.

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I. INTRODUCTION

While a sizeable electron-phonon coupling is prerequisite for polaron formation, it is not generally true that a system with strong electron-phonon interactions ($g>1$) has to display polaronic features. In a simple metal with wide bare bands and long-range forces like lead, for instance, intersite off-diagonal hopping terms prevent trapping of the charge carriers and onset of a polaronic state. In strong-coupling systems with a narrow electronic band D the antiadiabatic inequality $D/\hbar\bar{\omega}<1$ ($\bar{\omega}$ being the characteristic phonon frequency) is likely fulfilled thus ensuring the stability of the unit comprising electron and phonon cloud, that is, the small polaron.[1] Also in adiabatic regimes small polarons can form although a real-space broadening of the quasiparticle has to be expected, at least in three-dimensional systems. As a consequence, the adiabatic polaron mass renormalization is less strong than that occurring for the antiadiabatic polaron once the strength of the electron-phonon coupling has been fixed.[2] Theoretical investigation on these issues has become intense after that a polaronic mechanism has been proposed[3] as a viable possibility to explain high-T_c superconductivity with its peculiar transport properties. Evidence has been recently provided[4] for the polaronic nature of carriers also in perovskite manganites with colossal magnetoresistance (CMR) and a strong coupling of carriers to Jahn-Teller lattice distortions has been suggested[5] to explain the resistivity peak located at the Curie temperature.[6] Although polarons are invoked to account for different resistivity behaviors in systems with some lattice distortions,[7] the common origin, if any does exist, of such behaviors remains unexplained and a unifying theory is lacking. In general, a strong electron-phonon coupling implies violation of the Migdal theorem[8] and polaron collapse of the electron band with the appearance of time retarded interactions in the system. The path-integral method[9,10] has proved successful in dealing with this problem since a retarded potential naturally emerges in the exact integral action. Both the Fröhlich[11,12] and the Holstein[13–15] polaron properties have been studied by using path-integral techniques.

In this paper we propose a path-integral approach to the problem of a polaron scattered by a local lattice instability arising from an anharmonic phonon mode. Small polarons are assumed to exist by virtue of the strong *overall* electron-phonon coupling and independently of the *single* anharmonic mode. As an example, the latter could be associated with the motion of apical oxygen atoms in $YBa_2Cu_3O_7$ superconductor where a *c*-axis polarized high-frequency phonon couples to in-plane charge carriers.[16] On the other hand, the highly correlated motion of in-plane holes and *c*-axis lattice vibrations could in *itself* give origin to a small polaron.[17] We model the local instability by a double-well potential in its two state configuration, a two level system (TLS), and the one-dimensional atomic path between the two equilibrium minima is taken as time dependent.[18] Retardation effects are thus naturally introduced in the full partition function and the path-integral method permits us to derive the effective time (temperature)-dependent coupling strengths which control the resistivity.

II. PATH-INTEGRAL MODEL

Our starting point is contained in the following Hamiltonian:

$$H_0(\tau)=\bar{\epsilon}(g)\tilde{c}^{\dagger}(\tau)\tilde{c}(\tau)+\sum_{\mathbf{q}}\omega_{\mathbf{q}}a^{\dagger}_{\mathbf{q}}(\tau)a_{\mathbf{q}}(\tau)+H_{TLS}(\tau)$$

$$[H_{TLS}(\tau)]=\begin{pmatrix}0 & \lambda Q(\tau)\\ \lambda Q(\tau) & 0\end{pmatrix}$$

$$H_{int}(\tau)=-2\lambda Q(\tau)\tilde{c}^{\dagger}(\tau)\tilde{c}(\tau)$$

$$Q(\tau)=-Q_o+\frac{2Q_o}{\tau_o}(\tau-t_i) \qquad (1)$$

τ is the time which scales as an inverse temperature according to the Matsubara Green's-function formalism. $H_0(\tau)$ is the free Hamiltonian made of: (i) a polaron created (destroyed) by $\tilde{c}^{\dagger}(\tau)[\tilde{c}(\tau)]$ in an energy band $\bar{\epsilon}(g)$ whose width decreases exponentially by increasing the strength of the overall electron-phonon coupling constant g, $\bar{\epsilon}(g)=D\exp(-g^2)$; (ii) a diatomic molecular lattice whose phonon frequencies $\omega_{\mathbf{q}}$ are obtained analytically through a force

constant approach; (iii) a two level system in its symmetric ground-state configuration due to an anharmonic phonon mode. $Q(\tau)$ is the one-dimensional *space-time* hopping path followed by the atom which moves between two equlibrium positions located at $\pm Q_o$. τ_o is the bare hopping time between the two minima of the TLS and t_i is the instant at which the ith hop takes place. One atomic path is characterized by the number $2n$ of hops, by the set of t_i $(0<i\leqslant 2n)$ and by τ_o. We have implicitly assumed that the class of τ-linear paths yields the main contribution to the full partition function of the interacting system.[19] The closure condition on the path is given by: $(2n-1)\tau_s+2n\tau_0=\beta$, where β is the inverse temperature and τ_s is the time one atom is sitting in a well. The interaction is described by $H_{int}(\tau)$ with λ being the coupling strength between TLS and polaron, $\lambda Q(\tau)$ is the renormalized (versus time) tunneling energy which allows one to introduce the τ dependence in the interacting Hamiltonian.[20]

Following a method previously developed[19] in the study of the Kondo problem, we multiply $\lambda Q(\tau)$ by a fictitious coupling constant s $(0\leqslant s\leqslant 1)$ and, by differentiating with respect to s, one derives the one path contribution to the partition function of the system

$$\ln\left(\frac{Z(n,t_i)}{Z_0}\right)=-2\lambda\int_0^1 ds\int_0^\beta d\tau Q(\tau)\lim_{\tau'\to\tau^+} G(\tau,\tau')_s, \tag{2}$$

where Z_0 is the partition function related to H_0 and $G(\tau,\tau')_s$ is the full propagator for polarons satisfying a Dyson's equation:

$$G(\tau,\tau')_s=G^0(\tau,\tau')+s\int_0^\beta dy G^0(\tau,y)\lambda Q(y)G(y,\tau'). \tag{3}$$

The polaronic free propagator G^0 can be derived exactly in the model displayed by Eqs. (1). We get the full partition function of the system by integrating over the times t_i and summing over all possible even number of hops:

$$Z_T=Z_0\sum_{n=0}^{\infty}\int_0^\beta\frac{dt_{2n}}{\tau_0}\cdot\cdot\int_0^{t_2-\tau_0}\frac{dt_1}{\tau_0}\exp[-\beta E(n,t_i,\tau_0)]$$

$$\beta E(n,t_i,\tau_0)=Ł-(K^A+K^R)\sum_{i>j}^{2n}\left(\frac{t_i-t_j}{\tau_0}\right)^2 \tag{4}$$

with $E(n,t_i,\tau_0)$ being the one path atomic energy. Ł, which is a function of the input parameters, is not essential here while the second addendum in Eq. (4) is *not local in time* as a result of the retarded polaronic interactions between successive atomic hops in the double-well potential. K^A and K^R are the one path coupling strengths containing the physics of the interacting system. K^A (negative) describes the polaron-polaron attraction mediated by the local instability and K^R (positive) is related to to the repulsive scattering of the polaron by the TLS. Computation of $E(n,t_i,\tau_0)$ and its derivative with respect to τ_0 shows that the largest contribution to the partition function is given by the atomic path with $\tau_s=0$. The atom moving back and forth in the double well minimizes its energy if it takes the path with the highest τ_0 value allowed by the boundary condition, that is with $(\tau_0)_{max}=(2nK_BT)^{-1}$. This result, which is general, provides a criterion to determine the set of dominant paths for the atom at any temperature. Then, the effective interaction strengths $\langle K^A\rangle$ and $\langle K^R\rangle$ can be obtained as a function of T by summing over n the dominant paths contributions:

$$\langle K^A\rangle=-(\lambda Q_0)^2B^2\exp\left(2\sum_{\mathbf{q}}A_{\mathbf{q}}\right)\sum_{\mathbf{q}}A_{\mathbf{q}}\omega_{\mathbf{q}}^2\tilde{f},$$

$$\langle K^R\rangle=-\beta(\lambda Q_0)^3B^3\exp\left(3\sum_{\mathbf{q}}A_{\mathbf{q}}\right)\sum_{\mathbf{q}}A_{\mathbf{q}}\omega_{\mathbf{q}}^2\tilde{f} \tag{5}$$

with $B=[n_F(\overline{\epsilon})-1]\exp[-g\Sigma_{\mathbf{q}}\coth(\beta\omega_{\mathbf{q}}/2)]$ and $A_{\mathbf{q}}=2g\sqrt{N_{\mathbf{q}}(N_{\mathbf{q}}+1)}$.

$N_{\mathbf{q}}$ is the phonon occupation factor and $n_F[\overline{\epsilon}(g)]$ is the Fermi distribution for polarons. $\tilde{f}=\Sigma_{n=1}^{N}(\tau_0)_{max}^4$ and N is the cutoff on the number of hops in a path. The particular form of $(\tau_0)_{max}$ suggests that many hops paths are the relevant excitations at low temperatures whereas paths with a low number of hops provide the largest contribution to the partition function at high temperatures. Since the effective couplings which determine the resistivity depend on $(\tau_0)_{max}^4$, hence on N^{-3} (through $\tilde{f}$), a relatively small cutoff $(N\simeq 4)$ ensures numerical convergence of Eqs. (5) in the whole temperature range. On the other hand the non retarded term Ł in Eq. (4) has a slower $1/N$ behavior, therefore a larger cutoff should be taken at low temperatures where computation of equilibrium properties such as specific heat is strongly influenced by many hops atomic paths between the minima of the double-well excitations.

Our lattice Hamiltonian is made of diatomic sites whose intramolecular vibrations can favor trapping of the charge carriers.[13] The *intramolecular* frequency ω_0 largely influences the size of the lattice distortion associated with polaron formation[21] while the dispersive features of the phonon spectrum controlled by the *intermolecular* couplings are essential to compute the polaron properties both in the ground state and at finite temperatures. The range of the intermolecular forces is extended to the second neighbors shell since these couplings remove the phonon modes degeneracy (with respect to dimensionality) at the corners of the Brillouin zone thus permitting to estimate with accuracy the relevant contributions of high-symmetry points to the momentum space summations.

In a simple cubic lattice model with first and second neighbors molecular sites interacting via a force constants pair potential, the $\omega_{\mathbf{q}}$ are

$$\omega_{\mathbf{q}}^2=\frac{\alpha+3\gamma+4\delta}{M}+\frac{1}{M}\sqrt{\alpha^2+F_{x,y,z}}$$

$$\begin{aligned}F_{x,y,z}=&\gamma^2[3+2(c_xc_y+s_xs_y+c_xc_z+s_xs_z+c_yc_z+s_ys_z)]\\&+2\alpha\gamma(c_x+c_y+c_z)+4\alpha\delta(c_xc_y+c_yc_z)\\&+2\gamma\delta[3c_y+2c_z+c_x+2\cos(q_x-q_y-q_z)\end{aligned}$$

$$+2c_x(c_yc_z+s_xs_z)+c_xc_{2y}+s_xs_{2y}+c_yc_{2z}+s_ys_{2z}]$$
$$+2\delta^2[2+2c_xc_z+2c_z(c_xc_{2y}+s_xs_{2y})+c_{2y}+c_{2z}], \tag{6}$$

where $c_x=\cos q_x$, $s_x=\sin q_x$, $c_{2y}=\cos 2q_y$, etc. The intramolecular force constant α is related to ω_0 by $\omega_0^2=2\alpha/M$ with M being the reduced molecular mass. γ and δ are the intermolecular first and second neighbors force constants, respectively. Let us define $\omega_1^2=\gamma/M$ and $\omega_2^2=\delta/M$. Then the characteristic frequency $\overline{\omega}$, which we choose as the zone-center frequency, is: $\overline{\omega}^2=\omega_0^2+z\omega_1^2+z_{nnn}\omega_2^2$, z is the coordination number and z_{nnn} is the next-nearest-neighbors number.

We turn now to compute the electrical resistivity due to the polaronic charge carrier scattering by the impurity potential with internal degree of freedom provided by the TLS. Assuming *s*-wave scattering, one gets[22,8]

$$\rho=\rho_0\sin^2\eta,$$

$$\rho_0=\frac{3n_s}{\pi e^2 v_F^2 (N_0/V)^2\hbar}, \tag{7}$$

where n_s is the density of TLS which act as scatterers, v_F is the Fermi velocity, V is the cell volume, N_0 is the electron density of states, e is the charge, and $\hbar$ is the Planck constant. The phase shift η to the electronic wave function at the Fermi surface is related to the effective interaction strengths $\langle K^A\rangle$ and $\langle K^R\rangle$.

The input parameters of the model are six, that is, the three molecular force constants, g, D and the bare energy λQ_0. Q_0 can be chosen as $\simeq 0.05$ Å consistently with reported values in the literature on TLS's which are known to exist in glassy systems,[23,24] amorphous metals,[25] $A15$ compounds,[8] and likely in some cuprate superconductors.[17,26] In these systems the origin of the TLS's is not magnetic. The bare electronic band D is fixed at 0.1 eV. In Fig. 1, we take a rather large $\overline{\omega}$ setting the system in moderately adiabatic conditions and an intermediate ($g=1$) to very strong ($g>3$) electron-phonon coupling regime. The broad resistivity maximum developing at low temperatures in the intermediate regime clearly signals that the TLS's are at work here[27] while, by increasing T, the hopping time shortens and the incoming polarons cannot distinguish any more the TLS internal degree of freedom, then diagonal scattering prevails and a *quasilinear* resistivity behavior emerges at $T>200$ K. At larger g the polaron becomes progressively smaller in real space and the occurrence of the self-trapping event (at $g\simeq 3$) is marked by an abrupt increase in the effective mass, in any dimensionality.[28,29] As a consequence, the resistivity becomes nonmetallic and its absolute values are strongly enhanced. Let us fix $g=1$ which ensures polaron mobility and tune (see Fig. 2) the TLS-polaron coupling λ. We see that a resistivity peak located at $T\simeq 150$ K arises at $\lambda\geqslant 700$ meV Å^{-1} with height and width of the peak being strongly dependent on λ, and hence on the TLS energy. The low T resistivity still displays the maximum at the unitary limit while the high T ($T>300$ K) behavior can be metallic like ($\lambda<800$ meV Å^{-1}) or semiconducting like ($\lambda>800$ meV Å^{-1}). $\lambda\simeq 700$–800 meV Å^{-1} corresponds to a TLS energy of $\simeq 35$–40 meV which is comparable to the value of the bare polaron energy band $\overline{\epsilon}(g=1)\simeq 37$ meV. In this picture, the resistivity peak has a structural origin and it can be ascribed to resonant TLS-polaron scattering with effective attractive and repulsive interaction strengths becoming of the same order of magnitude at $T\simeq 150$ K. While the peak does not shift substantially by varying the strength of the intermolecular forces the height of the peak turns out to be rather sensitive to those strength, and hence to the size of the quasiparticle. As an example, by doubling ω_1 with respect to the value in Fig. 2 (at fixed ω_0 and ω_2), the normalized resistivity peak ($\lambda=1000$ meV Å^{-1} curve) drops to 0.65 while by taking $\omega_1=60$ meV the peak disappears. The effect of the second neighbors force constant is relatively

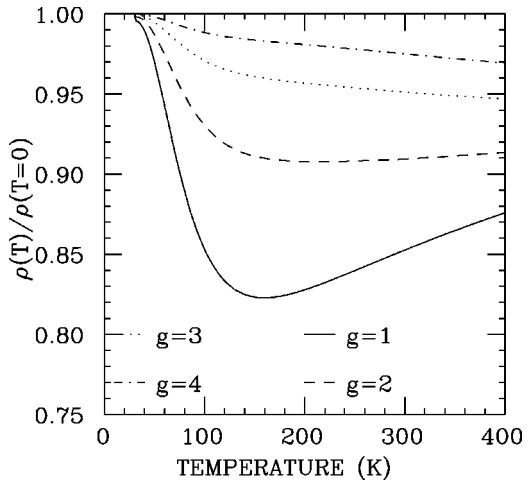

FIG. 1. Electrical resistivity normalized to the residual ($T=0$) resistivity for five values of electron-phonon coupling g. The bare TLS energy λQ_0 is 6.5 meV. The force constants which control the phonon spectrum are: $\omega_0=100$ meV, $\omega_1=20$ meV, $\omega_2=10$ meV.

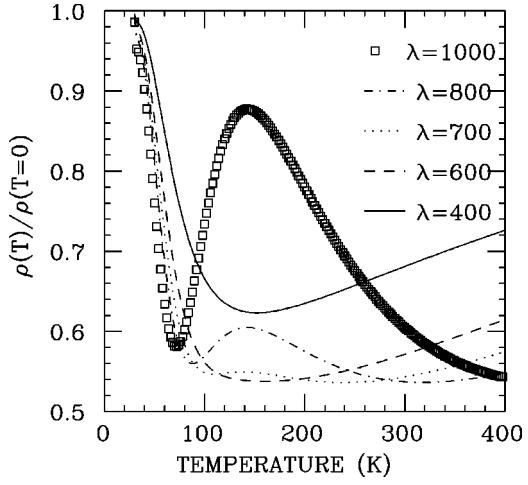

FIG. 2. Electrical resistivity normalized to the residual resistivity for five values of the polaron-TLS coupling λ. $g=1$. The intra- and intermolecular force constants are as in Fig. 1.

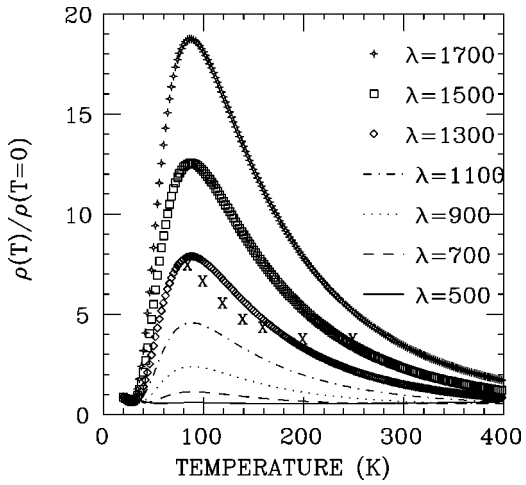

FIG. 3. Electrical resistivity normalized to the residual resisitivity for nine values of the polaron-TLS coupling λ. $g=1$, $\omega_0=50$ meV, $\omega_1=20$ meV, $\omega_2=10$ meV. The experimental data (X) are taken from Ref. 32.

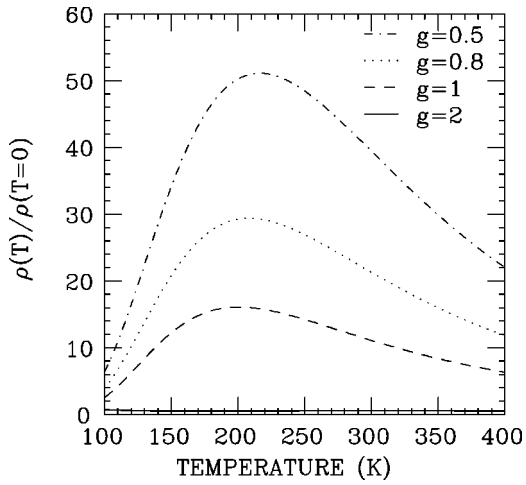

FIG. 4. Electrical resistivity normalized to the residual resistivity for four overall couplings g. $\lambda=5000$ meV Å^{-1}. $\omega_0=100$ meV, $\omega_1=80$ meV, $\omega_2=20$ meV.

less strong: by doubling ω_2 (at fixed ω_0 and ω_1) with respect to Fig. 2, the resistivity maximum becomes 0.76 while $\omega_2=30$ meV yields a peak value of 0.66.

At larger λ's the resonance peak is higher and broader since an increasing number of incoming polarons can be off diagonally scattered by the TLS. However, the appearance of this many-body effect mediated by the local potential does not change the position of the peak either, which instead can be shifted towards lower temperatures by reducing substantially ω_0, hence the $\overline{\omega}$ value. In Fig. 3, we set $\omega_0=50$ meV. As previously discussed[28] the intermolecular coupling ω_1 should be $\simeq\omega_0/2$ to ensure a correct application of the molecular lattice model with first neighbors interacting sites. Here we are extending the range of the forces to the second neighbors shell accordingly, ω_1 is slightly reduced to the 20-meV value while ω_2 is switched on and fixed at 10 meV. This choice of parameters allows us: (i) to treat correctly the ground-state polaron properties versus dimensionality,[28] (ii) to set the characteristic phonon frequency of the present three-dimensional lattice model at $\overline{\omega}\simeq 75$ meV which is the energy of *c*-axis polarized phonons due to apical oxygen vibrations coupled to the holes in the Cu-O planes of $YBa_2Cu_3O_{7-\delta}$.[16] Although our simple cubic lattice does not account for the details of the structural effects of YBCO we are in the appropriate range of parameters to capture the main features of the lattice polarons scattered by local instabilities in those compounds.

Anharmonic features of the oxygen modes have been recognized to be larger in underdoped samples[30] and doping dependent polaron formation[31] has been correlated to distortions of the oxygen environment. Our model predicts (Fig. 3) a large resisitivity peak whose height is strongly increased with respect to Fig. 2 (compare the $\lambda=700$ meV Å^{-1} cases) due to the fact that the polaron effective mass is heavier when lower energy phonons build up the quasiparticle. The nonmetallic behavior at T larger than $\simeq 90$ K reminds us of the anomalous *c*-axis resistivity observed in underdoped high-T_c superconductors.[32] We believe that the semiconductinglike ρ_c in underdoped high-T_c superconductors can be ascribed to anharmonic potentials due to oxygen displacements strongly coupled via λ (as in Fig. 3) to polaronic carriers. To attempt a comparison with experiments we need to fix the residual resistivity through the ground-state parameters given in Eq. (7). By extrapolating to $T=0$ the normal-state data on $YBa_2Cu_3O_{7-\delta}$ one derives $\rho(T=0)\simeq 3$ mΩ cm, hence the experimental peak value $\rho\simeq 20$ mΩ cm observed in the 80-K compound $YBa_2Cu_3O_{6.87}$ (Ref. 32) can be reproduced in my model by $\lambda\simeq 1300$ meV Å^{-1} which corresponds to a local mode energy of $\simeq 65$ meV, in fair agreement with the measured energies of phonons strongly coupled to the charge carriers.

In the range $80<T<200$ K the $\lambda\simeq 1300$ meV Å^{-1} curve fits rather well the data of Ref. 32. At $T=200$ K, the experimental ρ is $\simeq 10$ mΩ cm and the calculated value is $\simeq 8$ mΩ cm. In the range $200<T<300$ K, the experimental ρ is essentially flat while my results still exhibit a $d\rho/dT<0$ behavior. In general, the resistivities here obtained have an exponential temperature dependence (at T larger than the resonance) consistent with a polaronic hopping motion whereas the experimental data show a variety of sample dependent nonmetallic behaviors for the out-of-plane resistivity[33] in underdoped compounds.

A resistivity peak at $T\simeq 250$ K is observed in CMR materials[6] and a splitting of the Mn states due to a local lattice distortion has been proposed as a possible mechanism. In our description, only extended polarons dragging a cloud of high-frequency optical phonons (see Fig. 4) can produce a peak at $T\simeq 200$–250 K which is however broader than the experimental one. High phonon frequencies imply reduced mass renormalization and, consistently, weak overall electron-phonon coupling values. Extended polarons move therefore in wide energy bands and the resonance effect can take place only if the TLS splitting energy is large enough. In fact, by taking $D=0.5$ eV and $\lambda\simeq 5000$ meV Å^{-1} (Fig. 4), we get the peak at $g\leqslant 1$ whereas, in a small polaron regime ($g>1$) the resonance is lost and the peak is sup-

pressed. Although more specific models accounting for magnetic-field effects are required for the CMR systems, we argue that the strong coupling of polarons to structural distortions is likely relevant to their transport properties.

III. CONCLUSION

The path-integral method provides a powerful tool to describe *nonlocal (in time)* scattering of the charge carriers by *local (in space)* potentials due to structural distortions. Our charge carriers are lattice polarons which exist in the diatomic molecular lattice by virtue of a sizeable overall electron-phonon coupling. The structural instability is due to an anharmonic atomic motion between two equilibrium positions which typically could be $\simeq 0.1$ Å apart. I have derived the full partition function of the interacting system and obtained the temperature-dependent effective couplings by summing over the class of energetically favored atomic hopping paths. We suggest that the retarded nature of the interactions is the *key* ingredient to explain some anomalous transport properties observed in real materials. A resistivity peak is obtained as a consequence of resonant scattering of polarons strongly coupled to local double-well potentials.

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