Polaron formation as a genuine nonequilibrium phenomenon

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Solitons and polarons occurring in nonequilibrium steady states are investigated in the spinless Takayama Lin-Liu Maki (TLM) model. We propose the possibility of a new type of polarons in a nonequilibrium steady state. This is a genuine nonequilibrium phenomenon, since there is a threshold current below which they do not exist. It is considered to be an example of microscopic dissipative structure.

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Nearly 30 years ago, nonmetal-metal transition by doping halogens in polyacetylene was discovered.[1] The Su, Schrieffer, Heeger (SSH) model[2] and its continuous counterpart, the Takayama Lin-Liu Maki (TLM) model,[3] are known to describe many experimental results. Using these models, defects in polymers such as solitons and polarons have been extensively studied.[4–6] Subsequently, transport properties have been studied both experimentally and theoretically,[7] and it is known that charge-localized excitations such as solitons and polarons play an essential role in transport in polymers. In spite of these developments, there still remain several important issues to be settled.

Recently, the critical dopant concentration for the nonmetal-metal transition was improved[8] and the absence of the soliton contribution to the current was proposed.[9] However, the energetically most preferable state among those with solitons, polarons, bipolarons, and no localized excitations is not fully understood.[10,11]

The stability of polarons under various perturbations, such as an external electric field[12–15] and thermal noise[16,17] is still intensively studied even now, as is the dependence of the polaron velocity on the applied field[14,18,19] and/or Coulomb interaction.[20] Thus far, these theoretical works show only the *destructive* role of electric field and temperature on the stability of polarons. In this paper, we propose a *constructive* role for the current, namely, we show that current induces a new class of polarons within the TLM model. This is a genuine nonequilibrium property, since there exists a threshold current below which the polarons do not exist.

At equilibrium, polarons are induced by breaking the particle-hole symmetry, and they exist only for a spinful system. Experimentally, such polarons have been created by using photoinduced or doping techniques (see references of Refs. 7 and 21). The polarons that we shall discuss in this paper are induced by a different mechanism, and exist both for the spinful and spinless systems. Although our discussions are valid for both spinful and spinless systems, we discuss the *spinless* TLM model to emphasize the role of the current.

The Hamiltonian $H \equiv H_S + V + H_B$ is composed of H_S for the finite TLM chain, H_B for the reservoirs, and V for their interaction, which are given by

$$H_S = \int_0^\ell dx\Psi^\dagger(x)\left[-i\hbar v\sigma_y\frac{\partial}{\partial x} + \hat{\Delta}(x)\sigma_x\right]\Psi(x) + \frac{1}{2\pi\hbar v\lambda}\int_0^\ell dx\left[\hat{\Delta}(x)^2 + \frac{1}{\omega_0^2}\hat{\Pi}(x)^2\right]$$

$$V = \int d\boldsymbol{k}\,\{\hbar v_{\boldsymbol{k}} e^\dagger(0)a_{\boldsymbol{k}L} + \hbar v_{\boldsymbol{k}} d^\dagger(\ell)a_{\boldsymbol{k}R} + \text{(h.c.)}\} \quad (1)$$

$$H_B = \int d\boldsymbol{k}\,\hbar(\omega_{\boldsymbol{k}L}a^\dagger_{\boldsymbol{k}L}a_{\boldsymbol{k}R} + \omega_{\boldsymbol{k}R}a^\dagger_{\boldsymbol{k}R}a_{\boldsymbol{k}R}),$$

where $\Psi(x) = (d(x),e(x))^T$ is the two-component spinless fermionic field satisfying the boundary condition $d(0) = 0$, $e(\ell) = 0$; $\hat{\Delta}(x)$ is the lattice distortion; $\hat{\Pi}(x)$ is the momentum conjugate to $\hat{\Delta}(x)$; $a_{\boldsymbol{k}\nu}$ $(\nu = L,R)$ are the annihilation operators for reservoir fermions with wave number $\boldsymbol{k}$; $\hbar\omega_{\boldsymbol{k}\nu}$ represents their energies measured from the zero-bias chemical potential at absolute zero temperature; σ_x and σ_y are the Pauli matrices; ℓ is the length of the system; v is the Fermi velocity; λ is the dimensionless coupling constant; and ω_0 is the phonon frequency. We assume that the coupling matrix elements $v_{\boldsymbol{k}}$ as well as the density of states of the reservoirs are independent of energy;[22] thus, the integral

$$\frac{1}{i}\int d\boldsymbol{k}\frac{|v_{\boldsymbol{k}}|^2}{\omega - \omega_{\boldsymbol{k}\nu} - i0} \sim \pi\int d\boldsymbol{k}|v_{\boldsymbol{k}}|^2\delta(\omega - \omega_{\boldsymbol{k}\nu}), \quad (\nu = L,R)$$

becomes a positive constant Γ.

Next we describe the mean-field approximation, which is considered to be valid if the number of polymer chains is large.[21,23,24] Since we are interested in nonequilibrium steady states (NESS), the self-consistent condition is derived from the Heisenberg equation of motion for the lattice distortion,

$$\frac{\partial^2\hat{\Delta}(x,t)}{\partial t^2} = -\omega_0^2\{\hat{\Delta}(x,t) + \pi\hbar v\lambda\Psi^\dagger(x,t)\sigma_x\Psi(x,t)\}.$$

Namely, the self-consistent equation is written as

$$\Delta(x) + \pi\hbar v\lambda\langle\Psi^\dagger(x,t)\sigma_x\Psi(x,t)\rangle_\infty^{MF} = 0, \quad (2)$$

where $\Delta(x)$ is the mean-field NESS average of $\hat{\Delta}(x)$ and $\langle\cdots\rangle_\infty^{MF}$ represents the mean-field NESS average. The mean-field NESS corresponds to the initial state where two reservoirs are in equilibrium with different chemical potentials, and is characterized by the scattering theory of the NESS proposed by Ruelle.[25–27] Namely, the NESS is characterized as a state satisfying Wick's theorem with respect to the *incoming* fields $\alpha_{\boldsymbol{k}\nu}$ $(\nu = L,R)$ of the mean-field Hamiltonian, and having the two-point functions:[28,29]

$$\langle \alpha^{\dagger}_{\boldsymbol{k}\nu}\alpha_{\boldsymbol{k}'\nu}\rangle_{\infty} = f_{\nu}(\hbar\omega_{k\nu})\delta(\boldsymbol{k}-\boldsymbol{k}'), \quad (\nu = L,R) \tag{3}$$

where $f_\nu(x) \equiv 1/(\exp\{(x-\mu_\nu)/T\}+1)$ is the Fermi distribution function with temperature T, and chemical potentials $\mu_L = -eV/2$ and $\mu_R = eV/2$ (the Boltzmann constant is set to be unity). To be more concrete, expectation values of any observables at NESS can be calculated by rewriting observables in terms of the incoming field $\alpha_{k\nu}$ and then applying the Wick theorem and Eq. (3). The second term of Eq. (2) and the current Eq. (4) are calculated in such a way. (One can find similar calculations in Refs. 29 and 30). The current from the left to the right reservoir is calculated by counting the variation in the number of electrons in either reservoir, $\langle \frac{d}{dt}\int dk\, a^{\dagger}_{k\nu}a_{k\nu}\rangle^{MF}_{\infty}$:

$$J = \frac{G_0}{e}\int_{|\Delta_0|<|\epsilon|<\hbar\omega_c} d\epsilon \frac{\sqrt{\epsilon^2-\Delta_0^2}}{|\epsilon|}\,[f_R(\epsilon)-f_L(\epsilon)], \tag{4}$$

where $G_0 = e^2 v\Gamma/\{\pi\hbar(v^2+\Gamma^2)\}$ is the conductance in the normal phase. If there is sliding of the charge density wave (CDW), then the current given by Eq. (4) is considered to be the background current. (The possibility of sliding strongly depends on commensurability, the amount of impurities, and the electric field.)

We first briefly review our previous results[29] on the uniform solution $\Delta(x) = \Delta_0$, in which the fermionic spectrum has a gap $2|\Delta_0|$, and Δ_0 obeys the gap equation

$$\int_{|\Delta_0|/\hbar}^{\omega_c} d\omega \sum_{\nu=L,R} \frac{f_\nu(-\hbar\omega)-f_\nu(\hbar\omega)}{\sqrt{(\hbar\omega)^2-\Delta_0^2}} = \frac{2}{\hbar\lambda}, \tag{5}$$

where ω_c is the energy cutoff. This reduces to a well-known expression[3] at equilibrium in the absence of bias voltage. Equation (5) is valid when the chain length ℓ is sufficiently long. As is shown in Fig. 2, when $T < T^* \sim 0.5571 \times T_c$, we observe negative differential conductivity (NDC), which comes from the multivaluedness of the order parameter Δ_0 with respect to bias voltage. On the other hand, the order parameter is a single-valued function with respect to current. Thus, temperature and current are chosen as control parameters. The phase diagram on the J–T plane and the current dependence of the average

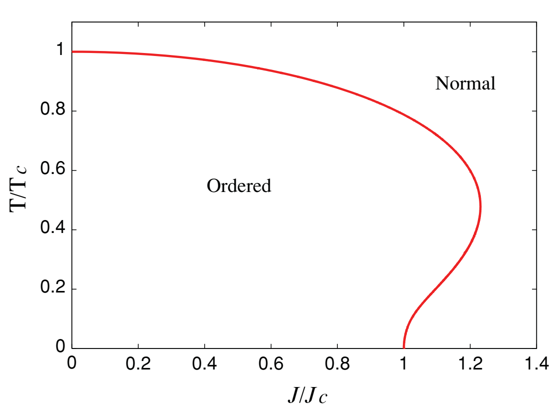

FIG. 1. (Color online) Phase diagram in the J-T plane.

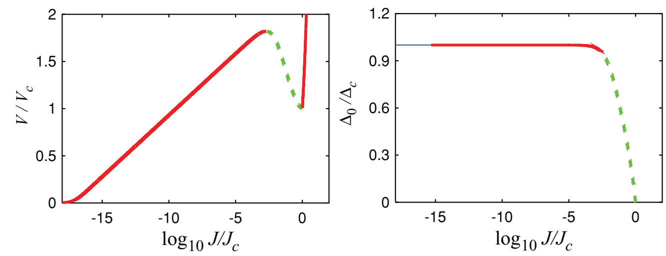

FIG. 2. (Color online) (Left) Current-voltage characteristics at $T = 0.05T_c$. (Right) Current dependence of $|\Delta_0|$ at $T = 0.05T_c$. In these figures, the solid line is stable and the dashed line is stable only at constant current. In the right figure, only the bold solid line admits polarons.

lattice distortion are shown, respectively, in Fig. 1 and 2, for $\lambda^{-1} = 2.4$.[31]

Among theoretical works, NDC was proposed in the field-driven SSH model,[32] as well as in the stochastically driven XXZ and extended Hubbard models.[33,34] Experimentally, both the I-V characteristic and the current dependence of the order parameter (Fig. 2) qualitatively agree with those of BEDT-TTF (Refs. 35 and 36). (They observe a current-induced suppression of the charge order. However, the mean-field TLM model and the mean-field extended Hubbard model are equivalent, and, as a consequence, our data qualitatively agree with these experimental data).

Next we investigate solitons and polarons. It is easy to verify that Eq. (2) admits a soliton solution[30] similar to that of the equilibrium case[3,37]:

$$\Delta(x) = \Delta_0 \tanh \kappa_s(x-a), \qquad \kappa_s = \frac{\Delta_0}{(\hbar v)},$$

where the amplitude Δ_0 is the solution of the gap equation (5) for the uniformly dimerized phase, and $a = O(\ell)$ represents the center of the soliton. At the same time, a midgap state appears with energy $\hbar\omega = 0$ in the fermionic spectrum. Then, following Brazovskii-Kirova[5] and Campbell-Bishop,[6] we look for a static polaron solution of the following form:

$$\Delta(x) = \Delta_0 - \hbar v\kappa_0(t_+ - t_-)$$

$$t_\pm \equiv \tanh \kappa_0(x-a\pm x_0), \quad \tanh 2\kappa_0 x_0 = \frac{\hbar v\kappa_0}{\Delta_0},$$

where Δ_0 and x_0 are parameters that are determined self-consistently, and a is the position of the polaron center of the order of ℓ. As in the equilibrium case, the corresponding fermionic spectrum consists of continuum states with energy $\hbar\omega = \pm\sqrt{(\hbar v k)^2+\Delta_0^2}$ $(|k| < \omega_c/v)$, and midgap states with energies $\hbar\omega = \pm\sqrt{\Delta_0^2-(\hbar v\kappa_0)^2} \equiv \pm\hbar\omega_B$. Even though the coupling between the midgap states and the reservoirs is exponentially small for long chain length ℓ, it still controls the occupation of the midgap states at NESS. Therefore, one should carefully take a long chain limit of the self-consistent Eq. (2), which will result in Eq. (6) (In the soliton case,[30] this exponentially small coupling was shown to give no contributions. On the other hand, it gives finite contributions in the polaron case, which makes a nonequilibrium polaron possible.):

$$
\begin{aligned}
I_B + I_S &= -\frac{\Delta(x)}{\hbar v \lambda} \\
I_S \equiv -\int_{|\Delta_0|/\hbar}^{\omega_c} d\omega &\frac{\omega^2 \Delta(x) - \omega_B^2 \Delta_0}{2\hbar v^2 \kappa \left(\omega^2 - \omega_B^2\right)} \frac{\sinh \hbar\beta\omega}{\cosh \hbar\beta\omega + \cosh \frac{\beta e V}{2}}, \\
I_B \equiv -\frac{\pi \omega_B}{4v}(t_+ - t_-) &\frac{\sinh \hbar\beta\omega_B}{\cosh \hbar\beta\omega_B + \cosh \frac{\beta e V}{2}},
\end{aligned}
\qquad (6)
$$

where $\beta = 1/T$, I_S is a contribution from the continuum states, and I_B is a contribution from the midgap states with energy $|\hbar\omega| < |\Delta_0|$ (see Ref. 30). Comparing term by term, the gap equation (5) is obtained, and the equation for energies $\pm\hbar\omega_B$ of the midgap states are

$$
\begin{aligned}
&\int_{|\Delta_0|/\hbar}^{\omega_c} \frac{\omega_B \, d\omega}{\sqrt{\omega^2 - \Delta_0^2/\hbar^2}\left(\omega^2 - \omega_B^2\right)} \frac{\sinh \hbar\beta\omega}{\cosh \hbar\beta\omega + \cosh \frac{\beta e V}{2}} \\
&\quad = \frac{\pi}{2v\kappa_0} \frac{\sinh \hbar\beta\omega_B}{\cosh \hbar\beta\omega_B + \cosh \frac{\beta e V}{2}}. \qquad (7)
\end{aligned}
$$

In this paper, we study pinned solitons and polarons, for which the current is still given by Eq. (4).

Equations (5) and (7) have a nontrivial solution *only* when the current (or, equivalently, the bias voltage) lies between the lower and upper threshold values $J_1(T) < J < J_2(T)$ ($V_1(T) < V < V_2(T)$), and the temperature is lower than T^*, below which the system shows NDC. As seen in Fig. 3, the polarons' width $2x_0$ and amplitude $A \equiv 2(\hbar v \kappa_0)^2/(|\Delta_0| + \hbar\omega_B)$ are decreasing functions of current. When the current (or bias voltage) approaches the lower threshold $J_1(T)$ ($V_1(T)$), the polarons' width diverges and their amplitude approaches the solitons' amplitude $2|\Delta_0|$. This indicates that the polarons split into soliton-antisoliton pairs. On the other hand, when the current (bias voltage) approaches the upper threshold $J_2(T)$ ($V_2(T)$), both the width and amplitude of the polarons vanish, and the polaron solution reduces to the uniform solution. Typical profiles of the polaron solution are shown in Fig. 3.

As mentioned above, $|\Delta_0|$ is a multivalued function of bias voltage and, for a given voltage, several uniform phases are possible. Although this suggests the possibility that collective local excitations can separate uniform domains with different values of $|\Delta_0|$, there exist only those interpolating uniform phases with the *same* $|\Delta_0|$, such as the solitons and polarons having the expressions we discussed. This is because charge conservation implies that the current J remains constant over the chain, and Δ_0 is a single-valued function of J. Also, it is interesting to note that the existence of the polaron solution is related to the linear stability studied previously.[29] Indeed, the polaron solution exists when the uniform phase with Δ_0 is stable *both* at constant current and constant bias voltage (the solid curves in the right figure of Fig. 2), but it does not exist if the uniform phase is unstable at constant voltage (the dashed curve in the right figure of Fig. 2). Because of this property, there is a one-to-one correspondence between current and bias voltage intervals where the polaron solution is possible [$J_1(T) < J < J_2(T)$ and $V_1(T) < V < V_2(T)$, respectively]. This aspect and the nonexistence of the polaron solution for $T > T^*$ deserve further investigation.

The possibility of the polaron solution at NESS can be qualitatively understood as follows. Recall that polarons at equilibrium are possible only in the spinful case. With the corresponding fermionic state, the lower midgap state is occupied by two fermions with opposite spins, and the upper midgap state is occupied by an unpaired fermion. In the half-filled spinless case at equilibrium, such an asymmetric occupation is not possible. This seems to suggest that it is necessary for the particle-hole symmetry to break for polaron formation. In contrast, at NESS, the particle-hole symmetry is broken by bias voltage even for the half-filled spinless case. This is because the fermionic occupation is controlled by $(f_L(\epsilon) + f_R(\epsilon))/2$, which is not symmetric under the exchange of particles and holes.

It is interesting to note that, at low temperatures, the width $J_2(T) - J_1(T)$ of current interval that admits polaron solutions increases with an increase in temperature, while the width $V_2(T) - V_1(T)$ of the voltage interval decreases. These behaviors of current and voltage are consistent, because the phases admitting a polaron solution tend to become insulating phases as $T \to 0$, which implies $\lim_{T\to 0}(J_2(T) - J_1(T))/(V_2(T) - V_1(T)) = 0$; thus, the decrease of $V_2(T) - V_1(T)$ with an increase of T does not contradict the increase of $J_2(T) - J_1(T)$. Because of the discontinuity of the RHS of Eq. (7) at $T = 0$, absolute zero temperature is a singular point if bias voltage is

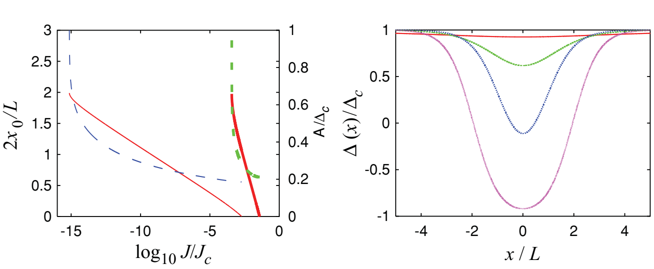

FIG. 3. (Color online) (Left) Current dependence of the amplitude A (the solid lines) and the soliton size $2x_0$ (the dashed lines) at $T = 0.05 \times T_c$ (thin lines) and $T = 0.2 \times T_c$ (bold lines). $2x_0$ is scaled by $L = \hbar v/\Delta_c$. (Right) A typical lattice profile at $T = 0.05 \times T_c$. From top to bottom, $J = 10^{-3} J_c$, $10^{-5} J_c$, $10^{-10} J_c$, and $10^{-15} J_c$.

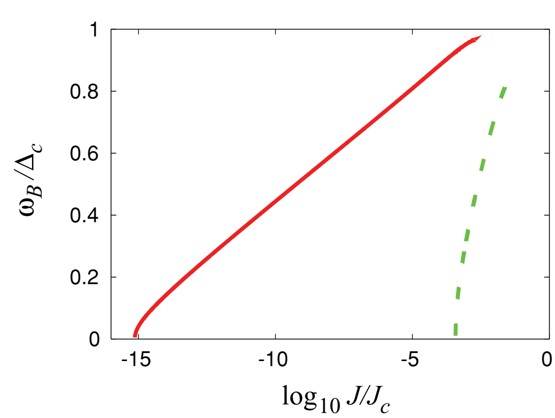

FIG. 4. (Color online) Current dependence of the positive-bound-state energy $\hbar\omega_B$ at $T = 0.05 \times T_c$ (solid line) and $T = 0.2 \times T_c$ (dashed line).

chosen as a control parameter. Indeed, at $T = 0$, Eqs. (5) and (7) admit a polaron solution with $\hbar\omega_B = (\pi^2/16 + 1)^{-1/2}\Delta_c$ only when $V = \{(\pi^2/16 + 1)\cosh^2\lambda^{-1}\}^{-1/2}\exp(\lambda^{-1})V_c$ and $J = 0$.

Experimentally, one of the most widely used tools to verify the existence of solitons and polarons has been spectroscopy, which allows to observe the energies of the associated midgap states.[7,38,39] It is thus interesting to study the current dependence of the energy of the midgap states. Figure 4 shows the current dependence of the energy $\hbar\omega_B$ for the midgap state at $T = 0.05 \times T_c, 0.2 \times T_c$ $(< T^*)$. As shown in the figure, $\hbar\omega_B$ is a monotonically increasing function of current, and it approaches 0 for $J \to J_1(T)$ and $|\Delta_0|$ for $J \to J_2(T)$; this reflects the change of the polarons' profile. Polarons in a *spinful* system possess the same properties, since the corresponding self-consistent equation is obtained simply by replacing λ in Eq. (5) with 2λ. Namely, the energies $\pm\hbar\omega_B$ of the midgap states associated with NESS polarons change from 0 to $\pm|\Delta_0|$ as current increases, while those with equilibrium polarons in a *spinful* system are fixed at $\pm\hbar\omega_B = \pm|\Delta_0|/\sqrt{2}$. Such a current-induced shift of energy spectra might be observed by spectroscopic experiments.

In summary, we have studied solitons and polarons in the open spinless TLM model, and specifically, we have shown that the current-induced polarons are possible only when currents above the critical value $J_1(T)$ [equivalently, a lower critical bias voltage $V_1(T)$] are applied to the system, and, thus, this polaron formation is a genuine nonequilibrium phenomenon. This observation suggests that this class of polaron is an example of microscopic dissipative structure. Also, we have shown that the critical temperature for the existence of a polaron solution is the same as the critical temperature for the appearance of NDC, although polarons do not appear with the currents of the NDC regime. The energies of the midgap states associated with polarons are shown to crucially depend on current, which might be observed by spectroscopic experiments.

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