PHYSICAL REVIEW B **78**, 241104(R) (2008)

Photoinduced Tomonaga-Luttinger-like liquid in a Mott insulator

Takashi Oka and Hideo Aoki
Department of Physics, University of Tokyo, Hongo, Tokyo 113-0033, Japan
(Received 8 September 2008; published 31 December 2008)

Photoinduced metallic states in a Mott insulator are studied for the half-filled one-dimensional Hubbard model with the time-dependent density-matrix renormalization group. An irradiation of strong ac fields is found to create, in the nonequilibrium steady state, a linear dispersion in the optical spectrum (current-current correlation) reminiscent of the Tomonaga-Luttinger liquid for the doped Mott insulator in equilibrium. The spin spectrum in nonequilibrium retains the des Cloizeaux–Pearson mode with the spin velocity differing from the charge velocity. The mechanism of the photocarrier doping, along with the renormalization in the charge velocity, is analyzed in terms of an effective Dirac model.

DOI: 10.1103/PhysRevB.78.241104 PACS number(s): 71.10.Fd, 71.30.+h, 72.20.Ht, 72.40.+w

I. INTRODUCTION

Doped one-dimensional (1D) Mott insulators are fascinating due to a special metallic state known as the Tomonaga-Luttinger (TL) liquid,[1] where excitations have collective nature as distinct from the conventional Fermi liquid.[2] Specifically, the charge velocity becomes renormalized due to the electron-electron interaction so that the charge excitation propagates with a velocity different from that of spin excitations—a hallmark of the "spin-charge separation." Experimentally, TL liquids have been observed in quantum wires and carbon nanotubes.[3]

Now, there is another way of making a Mott insulator metallic, which is entirely different from the chemical doping. Namely, photodoping is now being highlighted as a way to control the carrier density in 1D correlated systems.[4–10] Pump-probe experiments have shown that strong electric fields can turn Mott insulating crystals into metals.[4–6] Theoretically, the change in the conductivity by irradiation was studied in Refs. 9 and 10 with exact diagonalization. Features in the photodoping conceptually distinct from the chemical doping are (a) the system is out of equilibrium and (b) two types of carriers, i.e., electrons (equals as doubly occupied sites) and holes, coexist because they are pair produced. Although electron-hole systems in equilibrium have been studied in the past in terms of the TL theory,[11] properties, especially the spectral properties, of the *nonequilibrium* metallic states induced by irradiation are yet to be understood.

This has motivated the present work, where we shed light on this problem by studying the optical (current-current) and spin-correlation functions for a nonequilibrium steady state in 1D Mott insulators in strong ac electric fields. This is done in two steps: we first employ numerical simulations with the time-dependent density-matrix renormalization group (td-DMRG) (Ref. 12) to show that we have a TL-type linear dispersion in the photodoped system, where the charge and spin excitations have different velocities. We then confirm the result from a field-theoretic result. A starting point is in Refs. 13 and 14, where the present authors pointed out that there is an interesting analogy between the Schwinger mechanism in the decay of the quantum electrodynamics (QED) vacuum governed by the electron-positron creation rate[15] and the dielectric breakdown of Mott insulators in dc electric fields governed by the electron-hole creation rate. The corresponding phenomena in hadron physics is the formation of quark gluon plasma after a high energy impact of nuclei, where the quark anti-quark pair and the mass gap are the counterparts, respectively, of the electron hole pair and the Mott gap.[16] In one dimension, correlated electron systems can be mapped to a simple Dirac model via the massive Thirring model, which is dual to the sine-Gordon model for charge degrees of freedom of the 1D Hubbard model. We then analyze the photodoping with the effective Dirac model. There, the optical spectrum is formulated with the Floquet analysis for strong ac external fields. The result reproduces the main features in the td-DMRG result, providing a physical picture for the dynamics of the nonequilibrium collective modes in the photoinduced state.

II. NONEQUILIBRIUM STEADY STATE

We consider a Mott insulator in strong ac electric fields in the half-filled 1D Hubbard model with the Hamiltonian given, in standard notation, by $H(t)=H_0+H_F(t)$, where $H_0=-t_{\mathrm{hop}}\Sigma_{i\sigma}(c^{\dagger}_{i+1\sigma}c_{i\sigma}+\mathrm{H.c.})+U\Sigma_i n_{i\uparrow}n_{i\downarrow}$ and $H_F(t)=F\theta(t)\sin(\Omega t)\Sigma_i i n_i$ ($n_{i\sigma}=c^{\dagger}_{i\sigma}c_{i\sigma}$ and $n_i=n_{i\uparrow}+n_{i\downarrow}$). Here F and Ω are, respectively, the strength and frequency of the external electric field, which is switched on at $t=0$ (hence the insertion of a step function). In the calculation we take the length of the system $L=80$, a time step $\Delta t=0.04$, and the DMRG Hilbert-space size of $m=140$ in natural units. After obtaining the ground state $|\Psi_0\rangle$ of $H(t<0)$ with the finite-system DMRG algorithm,[18] we let the system evolve according to the time-dependent Hamiltonian $H(t)$ with the td-DMRG (Ref. 12) to obtain the wave function $|\Psi(t)\rangle=U(t;0)|\Psi_0\rangle$, with the time-evolution operator $U(t;t')=\hat{T}\exp(-i\int_{t'}^{t}H(s)ds)$ ($\hat{T}$ is the time ordering).

For $t>0$, the system relaxes into a nonequilibrium steady state, where we can define the photodoping rate as $x_{\mathrm{ph}}(t)=\frac{1}{L}\Sigma_i(\mathrm{av}\langle\Psi(t)|n_{i\uparrow}n_{i\downarrow}|\Psi(t)\rangle-\langle\Psi_0|n_{i\uparrow}n_{i\downarrow}|\Psi_0\rangle)$ which is the increment in the double occupancy. We also monitor the total energy, $E_{\mathrm{av}}(t)=\mathrm{av}\langle\Psi(t)|H(t)|\Psi(t)\rangle$. In these expressions, we eliminate the $T_{\mathrm{period}}=2\pi/\Omega$ oscillation by taking the average over each period, as denoted by "av." The time profiles of the photodoping rate and the total energy in Fig. 1 are similar, which indicates that the energy absorbed from the ac field is used to excite electron-hole pairs, i.e., photodoping. The sys-

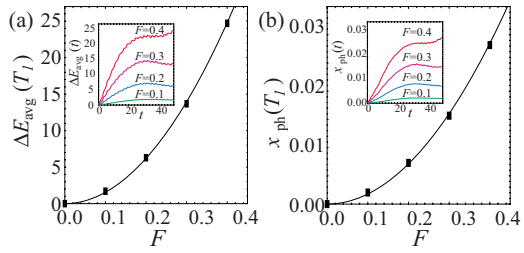

FIG. 1. (Color online) Inset: time-dependent DMRG result for the (a) total energy and (b) photodoping rate for the half-filled 1D Hubbard model with $U/t_{\rm hop}=8.0$ and $\Omega=8.0$. The main panels depict their final values at $t=T_1$ vs the field strength F, where the solid lines are parabola fitted to the data.

tem relaxes to a steady state until the situation where pair production rate=annihilation (stimulated emission) rate is achieved.

The doping rate depends both on the frequency Ω and the strength F of the electric field. When Ω is greater than the Mott gap Δ, excitations occur via one-photon absorptions. Even when $\Omega<\Delta$, multiphoton processes can excite the system above the gap for sufficiently strong fields, where the process becomes resonant when $m\,\Omega=\Delta$ with m being an integer.[19] The extreme case is the dc limit $\Omega\to 0$, where many-body Landau-Zener tunneling across the Mott gap induces metallization.[13,14,17]

III. CORRELATION FUNCTIONS

To characterize the 1D many-body system we calculate the correlation functions after a steady state is attained,

$$\chi_{AB}(t,T_1;i,j)=\langle\Psi(t)|A_i U(t,T_1)B_j|\Psi(T_1)\rangle, \qquad (1)$$

where A and B are operators, e.g., the current $J_i=-it_{\rm hop}\Sigma_\sigma(c^\dagger_{i+1\sigma}c_{i\sigma}-{\rm H.c.})$ or the spin $\vec{s}_i=\frac{1}{2}\Sigma_{\alpha\beta}c^\dagger_{i\alpha}\vec{\sigma}_{\alpha\beta}c_{i\beta}$, and T_1 is the time around which the steady state is reached (and the curves in Fig. 1 flatten; typically $T_1=50$ and 100 depending on the field strength). Physically, χ_{JJ} represents the probing process in standard pump-probe experiments, where a photon in the probe light generates a local electron-hole pair at position j. To focus on the interplay between the charge and spin degrees of freedom, we can compare in Fig. 2 the current correlation function with the behaviors of spin and charge. For the spin we define the local spin energy, $\varepsilon_i^{\rm spin}(t)/J_{\rm exch}\equiv\langle\Psi^J(t)|\vec{s}_{i+1}\cdot\vec{s}_i|\Psi^J(t)\rangle$, which measures the exchange energy (normalized by the exchange coupling $J_{\rm exch}=4t_{\rm hop}^2/U$).[20] For the charge we examine the double occupancy defined by $n_i^d(t)=\langle\Psi^J(t)|n_{i\uparrow}n_{i\downarrow}|\Psi^J(t)\rangle$. Here $|\Psi^J(t)\rangle=U(t,T_1)J_j|\Psi(T_1)\rangle$ is the state where a perturbation (J_j) is added at site j on the steady nonequilibrium state. The behavior of the three quantities shows that the temporal evolution after the probe excitation on j at $t=T_1$ propagates in two processes. The first is *diffusion of the doublon-hole pair*, which is followed by the *relaxation* process. The relaxation is seen as a decay of the current correlation, which is seen to be accompanied by a disturbance in the spin structure (as marked with a red broken line in the figure). This indicates that the spin and charge degrees of freedom become coupled

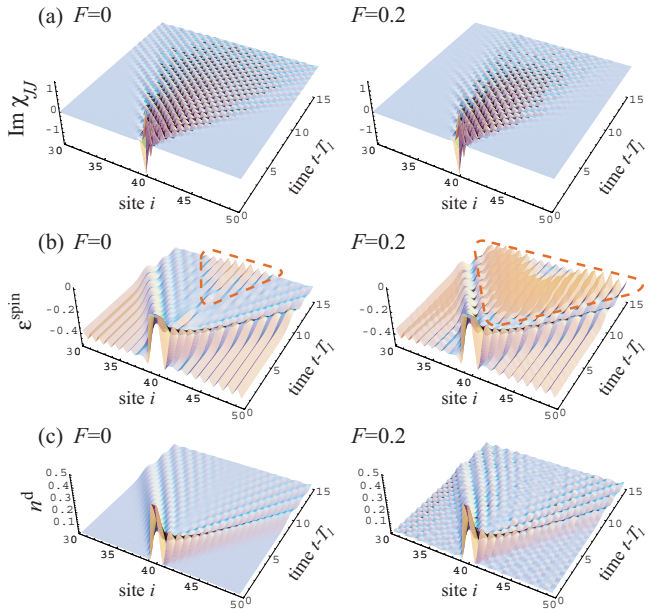

FIG. 2. (Color online) (a) Current-current correlation Im $\chi_{JJ}(t,T_1;i,j)$, (b) local spin energy $\varepsilon_i^{\rm spin}(t)$, and (c) double occupancy $n_i^d(t)$ for zero field $F=0$ (left panels) and for a finite field $F=0.2$ (right) with $U/t_{\rm hop}=8.0$ and $\Omega/t_{\rm hop}=8.0$. Here the probed position j in the pump-probe process is set to be at the center ($j=40$) of the 1D system with length of 80. Broken lines indicate the region for relaxation due to spin (see text). The small ripples [(c) right panel] are the $T=2\pi/\Omega$ period fluctuation.

more strongly, where spins act as a kind of energy reservoir for charges. Spin-charge coupling already exists in equilibrium for higher-energy states,[21] which is natural since charge excitations act as boundary conditions to spins with spin vanishing at doubly occupied or empty sites. The decay of the current correlation, already present for zero ac electric field ($F=0$), becomes faster in finite fields, which implies that the spin-charge coupling becomes stronger, due to higher-energy states becoming involved. However, in the field range studied here, the coupling is not strong enough to destroy the spin-charge separation picture,[10] i.e., the spin and charge degrees of freedom still have independent dispersions, as will be discussed below.

IV. COLLECTIVE EXCITATIONS IN NONEQUILIBRIUM

So what is the nature of the photoinduced carriers? We can obtain the excitation spectrum as the Fourier transform of the correlation functions,

$$\chi_{AB}(q,\omega)=\int_{T_1}^{T_1+T_2}dt\sum_j e^{i\omega(t-T_1)-iq(j-j_c)}\,{\rm Im}\,\chi_{AB}(t,T_1;j,j_c), \qquad (2)$$

where T_2 is set to be prior to the time at which the wave front reaches the sample boundary. We call the quantity, following the equilibrium case, the optical spectrum for χ_{JJ} and the spin spectrum for χ_{ss}.

A striking feature in the result in Fig. 3, a key finding here, is that while we have an optical gap (equals as a Mott

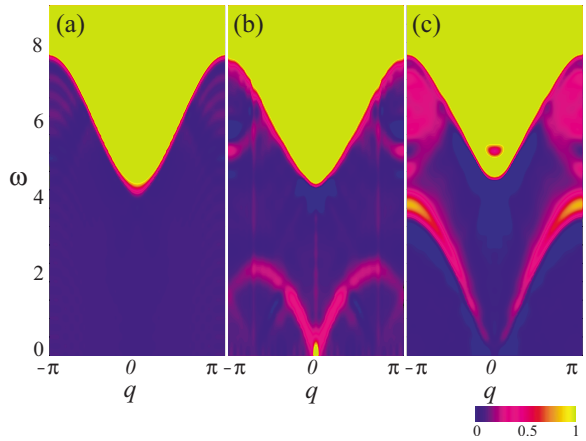

FIG. 3. (Color online) The optical spectrum, Im $\chi_{JJ}(q,\omega)$, with $U/t_{\mathrm{hop}}=8.0$ for (a) the half-filled case with no ac fields, (b) the half filled with a finite ac field $F/t_{\mathrm{hop}}=0.1$, and (c) the electron doped with no ac fields ($n=1.025, F=0$).

gap) in the zero ac field [Fig. 3(a)], a set of states with a zero-gap linear dispersion, $\omega_q \sim v_{\mathrm{PTL}}|q|$, emerges in the gap in finite ac fields [Figs. 3(b) and 3(c)]. We call the metallic state a *photoinduced Tomonaga-Luttinger-type liquid* in the sense that its charge velocity v_{PTL} is renormalized by the electron interaction. Its value is, in fact, similar to the equilibrium counterpart as can be seen by comparing the slope in nonequilibrium with that for the equilibrium-doped systems [Fig. 3(c)].

If we turn to the spin spectrum in Fig. 4, we can clearly see the des Cloizeaux–Pearson mode in equilibrium (precisely speaking, this was obtained by the Bethe ansatz method[22]), which survives in finite fields. We note that the charge and spin velocities defined and numerically obtained in nonequilibrium here are different. So in this particular sense we have a spin-charge separation. As the field becomes

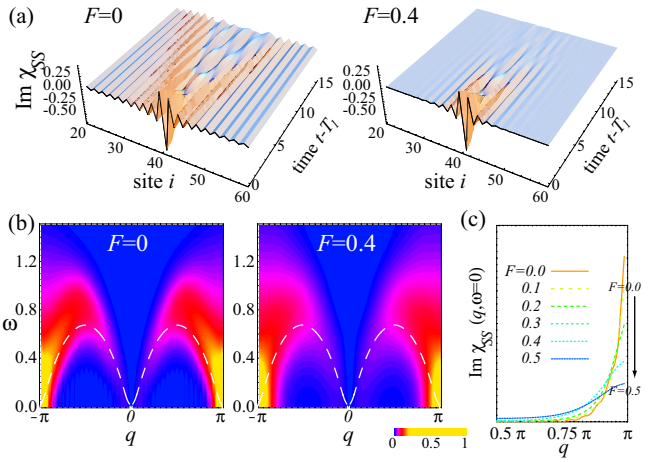

FIG. 4. (Color online) (a) Spin correlation Im $\chi_{ss}(t,T_1;i,j)$ for a zero field (left) and for a finite field $F=0.4$ (right) with $U/t_{\mathrm{hop}}=8.0$, $\Omega/t_{\mathrm{hop}}=8.0$. (b) Color-coded spin spectrum Im $\chi_{ss}(q,\omega)$ in the half-filled Hubbard model with $U/t_{\mathrm{hop}}=8.0$ for a zero ac field (left) or for a finite ac field $F/t_{\mathrm{hop}}=0.4$ (right). The white dashed lines are the des Cloiseaux–Pearson mode. (c) The AF peak Im $\chi_{ss}(q,\omega=0)$ vs q are also displayed for various field strengths.

stronger, the spectrum becomes blurred. Specifically, the antiferromagnetic (AF) fluctuation represented by the peak value $\chi_{ss}(q\sim\pi,\omega\sim 0)$ becomes smaller, which should be because the induced carriers act to melt the magnetic order.

V. EFFECTIVE MODEL

Due to its many-body nature, it is difficult to obtain a mathematically rigorous analysis of the photoinduced metallic state in Mott insulators. However, we can carve out some of its properties in terms of an effective model, which was initiated by Mori and Fukuyama,[23] Luther and Emery,[24,25] and Giamarchi[26] to study linear responses in the presence of a charge gap. In this approach we start from the 1+1-dimensional massive Thirring model. The charge degrees of freedom are represented by two spinless fermions $\Psi=[\psi_1(x),\psi_2(x)]$, where ψ_1 and ψ_2 represent left mover and right mover, respectively, and the Hamiltonian reads

$$H_\rho^{\mathrm{MT}}(t)=v_c\int dx\left[\Psi^\dagger(x)\left(-i\nabla_x(t)\sigma_3+\frac{\Delta}{2}\sigma_1\right)\Psi(x)\right]+H_I, \tag{3}$$

where σ_i is Pauli matrices, v_c is renormalized charge velocity, and Δ is the umklapp-scattering coupling constant, an ascendant of the original Mott gap at half filling. This model is dual to the sine-Gordon model, where the size of the interaction term $H_I=g\int dx[(\Psi^\dagger\Psi)^2-(\Psi^\dagger\tau_1\Psi)^2]$ translates to the sine-Gordon coupling through a duality relation.[27] However, in the following we make a further simplification, namely, we neglect the self-interaction H_I. This by no means implies a neglect of the original electron-electron interaction but amounts to neglect self-energy corrections to quasiparticle lifetime, etc. We employ this approximation to focus on the effect of the charge gap, which primarily appears from the first term in Eq. (3).

The ac electric field is taken as the coupling, $\nabla_x(t)=\partial_x+iA_1(t)$, in the above, with $A_1(t)=(F/\Omega)\sin\Omega t$. We then obtain the nonlinear and nonequilibrium evolutions of the state in this model. After a Fourier transform, the equation of motion becomes $i\frac{d}{dt}|\Psi_k(t)\rangle=[v_c[k+(F/\Omega)\sin\Omega t]\sigma_3+\frac{\Delta}{2}\sigma_1\}]|\Psi_k(t)\rangle$. Here we adopt the Floquet method for treating systems in ac fields (see, e.g., Ref. 28), i.e., we seek a solution of the form $|u_\alpha(k;t)\rangle=\Sigma_m e^{-i\varepsilon_\alpha(k)t-im\Omega t}|u_\alpha^m(k)\rangle$, where m is the Floquet index, α labels the eigenstate, and ε_α is the Floquet quasienergy. From the equation of motion, the Floquet modes satisfy a set of linear relations $\Sigma_n(H_{\mathrm{eff}})^{mn}|u_\alpha^n(k)\rangle=\varepsilon_\alpha(k)|u_\alpha^m(k)\rangle$, where the effective Floquet Hamiltonian has matrix elements $(H_{\mathrm{eff}})^{mm}=v_ck\sigma_3+\frac{\Delta}{2}\sigma_1-m\Omega\sigma_0$, $(H_{\mathrm{eff}})^{m,m\pm1}=\mp\frac{i}{2}v_c\frac{F}{\Omega}\sigma_3$. We consider a sudden switch on of the ac field, i.e., the time evolution starts from the ground state $|\psi_0(k)\rangle$ of $v_ck\sigma_3+\frac{\Delta}{2}\sigma_1$. The solution is then $|\psi_0(k;t)\rangle=\Sigma_\alpha\phi_\alpha(k)|u_\alpha(k;t)\rangle$ with $\phi_\alpha(k)=\langle u_\alpha^0(k)|\psi_0(k)\rangle$. The current correlation function in this model is[29]

$$\mathrm{Im}\,\chi^{\mathrm{Dirac}}(q,\omega)$$
$$=2e^2\int\frac{dk}{2\pi}\sum_{\alpha\beta m}[|\phi_\beta(k)|^2-|\phi_\alpha(k+q)|^2]\delta[\varepsilon_\beta(k)-\varepsilon_\alpha(k+q)+\omega]$$

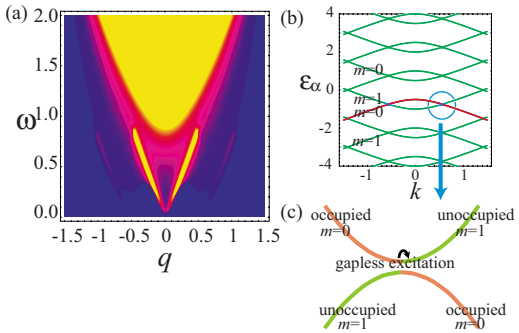

FIG. 5. (Color online) (a) The optical spectrum Im $\chi^{\mathrm{Dirac}}(q,\omega)$ of the effective Dirac model in a finite ac field with $\Omega/\Delta=1.5$ and $F=0.1$. (b) The Floquet quasienergy $\varepsilon_\alpha(q)$, where the red lines represent occupied states and green lines represent unoccupied states. (c) A schematic level repulsion.

$$\times \mathrm{Tr}[|u_\alpha^0(k+q)\rangle\langle u_\alpha^m(k+q)|\tau_3|u_\beta^m(k)\rangle\langle u_\beta^0(k)|\tau_3]. \qquad (4)$$

The optical spectrum [Fig. 5(a)] shows that a metallic linear-dispersion mode emerges in the gap in the presence of an ac field. The result does resemble the present td-DMRG result (Fig. 3). The spectrum, despite being nonequilibrium, is also similar to the massive Thirring model result for the doped 1D Mott insulator.[23] The origin of the gapless excitation in nonequilibrium can be traced back, in the present massive Thirring+Floquet analysis, to the quasienergy-level scheme in Fig. 5(b). In the absence of ac fields, the Dirac model has two branches with the lower band completely filled. As we turn on the ac field, we have a series of replicas (i.e., Floquet modes equally spaced by Ω) for each of the electron and hole branches. Physically, these modes correspond to m-photon absorbed states. As in standard quantum mechanics, level repulsion takes place when two modes cross with each other with a nonzero matrix element between them. In our model, the most important feature is the level repulsion between an occupied Floquet mode [red in Fig. 5(b)] and an unoccupied one (green). As shown in the blowup [Fig. 5(c)], gapless excitations then emerge across the occupied and unoccupied states, which should contribute to the optical spectrum when we evaluate Eq. (4). Specifically, the linear dispersion of the photoinduced state is $\omega_q \sim v_c|q|$, where the renormalized velocity v_c, a parameter independent of the spin velocity $v_s \propto t_{\mathrm{hop}}^2/U$, is conceived to have a value that depends on the detail of the irradiation.

Another interesting point is when the photon energy Ω is below the gap Δ. In this situation, multiphoton processes with $m \geq 2$ are necessary to excite the system. We can indeed show that, when the condition $\Omega \sim \Delta/m$ is met, the excitation becomes resonant and carriers are injected efficiently, which will be described elsewhere.

In conclusion, we have shown that a Mott insulator irradiated by strong ac electric fields has a collective mode which is reminiscent of the Tomonaga-Luttinger liquid in equilibrium. While spins and charges are coupled in the relaxation process, the collective modes have different spin and charge velocities. Emergence of the linear collective mode is also supported by an effective Dirac model.

ACKNOWLEDGMENTS

We have greatly benefited from discussions with Naoto Tsuji on the Floquet method. T.O. acknowledges Keiji Saito and Keiichiro Nasu for valuable discussions. This work has been supported in part by a Grant-in-Aid for Scientific Research on a Priority Area "Anomalous quantum materials" from the Ministry of Education, Science, Sports, and Culture of Japan.

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