Optical response of highly excited particles in a strongly correlated system

Zala Lenarčič,[1] Denis Golež,[1] Janez Bonča,[1,2] and Peter Prelovšek[1,2]
[1]*J. Stefan Institute, SI-1000 Ljubljana, Slovenia*
[2]*Faculty of Mathematics and Physics, University of Ljubljana, SI-1000 Ljubljana, Slovenia*
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We present a linear-response formalism for a system of correlated electrons out of equilibrium, as relevant for the probe optical absorption in pump-probe experiments. We consider the time dependent optical conductivity $\sigma(\omega,t)$ and its nonequilibrium properties. As an application we numerically study a single highly excited charged particle in the spin background, as described within the two-dimensional t-J model. Our results show that the optical sum rule approaches the equilibriumlike one very fast; however, the time evolution and the final asymptotic behavior of the absorption spectra in the finite systems considered still reveal dependence on the type of initial pump perturbation. This is observed in the evolution of its main features: the midinfrared peak and the Drude weight.

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I. INTRODUCTION

Time-resolved pump-probe optical spectroscopy represents a new powerful tool to study materials with strongly correlated electrons and offers a direct approach to far-from-equilibrium phenomena, probing in particular the relaxation and thermalization processes [1–6]. It appears that strongly correlated systems are in general characterized by very fast relaxation processes, emerging from the inherent strong interactions. Theoretical studies of nonequilibrium dynamics and transient phenomena in correlated models confirm this, even in particular states as the Mott-Hubbard insulator [7–9].

In theory, probing the transient nonequilibrium state with a weak electromagnetic pulse naturally leads to the linear-response approach. The optical conductivity $\sigma(\omega,t)$ is the time dependent dynamical quantity directly relevant to pump-probe optical spectroscopy and represents the response to the probing electric field in a nonequilibrium situation. Some care is needed to define and properly extract the sum rules and a possible dissipationless component—Drude weight (charge stiffness). Recently, such a formalism has been proposed for continuous correlated systems [10]. Adapted for the application of the dynamical mean-field theory (DMFT), it has been used for the analysis of the Hubbard model [8,11,12] or with the emphasis on the description of time dependent photoemission spectroscopy [13]. Another definition has been used to examine the dynamics of the Hubbard-Holstein model [14]. A stationary response, as a characteristic of a nonequilibrium quenched state, has also been considered recently and studied explicitly for a hard-core boson model [15] and in connection with the fluctuation-dissipation relation [16,17]. There are also studies in which the effect of the probe field after the intensive pulse excitation is directly followed by introduction of the (classical) driving time dependent electric field [18].

The aim of this paper is to introduce the differential optical conductivity $\sigma(\omega,t)$ representing the causal linear response of the lattice current to an arbitrary electric-field pulse $\mathbf{E}(t' > t)$, acting on a general nonequilibrium many-body wave function $|\psi(t)\rangle$ within a tight-binding model of correlated electrons. Such a formulation allows the definition and the consideration of the optical sum rule at any time, as well as the possible existence of the Drude weight $D(t)$ as the dissipationless response.

As a nontrivial example we test the formalism by numerical investigation of a single highly excited charged particle (hole) within Mott-Hubbard insulator, as represented by the two-dimensional (2D) t-J model. It seems plausible that, in the long-time and thermodynamic limit, anomalous $\sigma(\omega,t)$ should approach the ground state (g.s.) response $\sigma_0(\omega)$, since the particle is expected to relax to the g.s. by emitting the extra energy via relevant bosonic excitations, i.e., via magnons. On the other hand, in a closed finite system one would expect the response to approach a thermal equilibrium response $\sigma_{\rm th}(\omega)$, characterized by $T > 0$. Our numerical solutions indicate that these presumptions are only partly realized for concrete examples and that for the long-time response also the initial state plays a role, clearly visible at least within the limitation of our finite systems and finite evolution times.

The paper is organized as follows. In Sec. II we present the linear response formalism for the optical conductivity $\sigma(\omega,t)$ within the tight-binding model for the general nonequilibrium many-body wave function and related density matrix. Since the calculation of the time dependent $\sigma(\omega,t)$ in principle involves a two-time evolution, implementation with a single-time evolution is developed in order to reduce the numerical complexity as described in Sec. III. Section IV is devoted to the numerical study of a nontrivial test case, representing the optical response of the excited particle in the strongly correlated background, as given by the t-J model with a single hole. Conclusions and open questions are discussed in Sec. V.

II. NONEQUILIBRIUM OPTICAL LINEAR RESPONSE

In a general single-band tight-binding model of correlated electrons (with charge e), assuming the system with periodic boundary conditions (PBCs), the action of the electromagnetic field can be introduced via the vector potential $\mathbf{A}(t)$ through the usual gauge (Peierls) construction. The latter neglects the interband tunneling in multiorbital models and the field-induced distortions of the orbitals, but remains appropriate for the single-orbital case and weak fields. We consider the

tight-binding Hamiltonian (using $\hbar = 1$) up to $\mathcal{O}(\mathbf{A}^3)$:

$$H[\mathbf{A}(t)] = -\sum_{i,j,s} t_{ij} \exp[ie\mathbf{A}(t)\cdot\mathbf{R}_{ij}]c^\dagger_{js}c_{is} + H_{\rm int}$$
$$\approx H_0 - e\mathbf{A}(t)\cdot\mathbf{j} + \frac{e^2}{2}\mathbf{A}(t)\cdot\tau\mathbf{A}(t), \quad (1)$$

written with the particle current $\mathbf{j}$ and the stress tensor τ operators:

$$\mathbf{j} = i\sum_{i,j,s} t_{ij}\mathbf{R}_{ij}c^\dagger_{js}c_{is}, \quad \tau = \sum_{i,j,s} t_{ij}\mathbf{R}_{ij}\otimes\mathbf{R}_{ij}c^\dagger_{js}c_{is}, \quad (2)$$

where $\mathbf{R}_{ij} = \mathbf{R}_j - \mathbf{R}_i$. The electrical current

$$\mathbf{j}_e(t) = -\partial H/\partial\mathbf{A}(t) = e\mathbf{j} - e^2\tau\mathbf{A}(t) \quad (3)$$

is a sum of the particle current and the diamagnetic contribution. We treat the case in which the unperturbed system is described by a pure (nonequilibrium) many-body wave function (w.f.) $|\psi(t)\rangle$, with the time evolution operator $U(t',t) = \exp[-iH_0(t'-t)]$ corresponding to the time independent H_0. We use the standard formalism [19] to evaluate the linear response of $\langle\mathbf{j}\rangle_{t'}$ to a general $\mathbf{A}(t'')$ applied at t, whereas the diamagnetic part is already linear in $\mathbf{A}(t')$. Introducing the notation for expectation values $\langle O\rangle_t = \langle\psi(t)|O|\psi(t)\rangle$ and the interaction representation $B^I(t') = U^\dagger(t',t)BU(t',t)$,

$$\langle\mathbf{j}_e\rangle_{t'} = e\langle\mathbf{j}\rangle_{t'} - e^2\mathbf{A}(t')\langle\tau\rangle_{t'} + e^2\int_t^{t'} dt''\chi(t',t'')\mathbf{A}(t''),$$
$$\chi(t',t'') = i\theta(t'-t'')\langle[\mathbf{j}^I(t'),\mathbf{j}^I(t'')]\rangle_t. \quad (4)$$

Differential conductivity $\sigma(t',t)$ is defined through the response to electric field $\mathbf{E}(\bar{t})$:

$$\delta\langle\mathbf{j}_e\rangle_{t'} = V\int_t^{t'} d\bar{t}\,\sigma(t',\bar{t})\mathbf{E}(\bar{t}), \quad (5)$$

V being the volume of the system. Taking into account $\mathbf{A}(t'') = -\int_t^{t''}\mathbf{E}(\bar{t})d\bar{t}$ and Eqs. (4) and (5) we get

$$\sigma(t',t) = \frac{e^2}{V}\left[\langle\tau\rangle_{t'} - \int_t^{t'} dt''\chi(t',t'')\right]. \quad (6)$$

For a nonstationary state there is no unique definition of the frequency-dependent $\sigma(\omega,t)$ [8,10,15]. We choose a plausible relation reflecting the causality and switching on of the field at time t, i.e., $\mathbf{E}(\bar{t} < t) = 0$:

$$\sigma(\omega,t) = \int_0^{t_m} ds\;\sigma(t+s,t)e^{i\omega s}, \quad (7)$$

where t_m is the width of window in which we do the Fourier transformation, so that formally $t_m \to \infty$ but is in practice the maximum time of probe duration. With such a definition, Eq. (7), we avoid the ambiguity of including times prior to the pump pulse. The sum rule for so defined $\sigma'(\omega,t) = \mathrm{Re}\,\sigma(\omega,t)$ then follows directly from Eq. (7):

$$\int_{-\infty}^{\infty} d\omega\,\sigma'(\omega,t) = \pi\sigma(t,t) = \frac{\pi e^2}{V}\langle\tau\rangle_t. \quad (8)$$

It is evident that the sum rule, Eq. (8), is a time dependent quantity, i.e., $\langle\tau\rangle$ evaluated at the time t when the probe field is applied. Moreover, independent of the precise form of the Fourier transform it remains proportional to the $\langle\tau\rangle$ at the time held fixed in the transformation.

One can define also the Drude weight $D(t)$ as the dissipationless component:

$$\sigma'(\omega,t) = 2\pi e^2 D(t)\delta(\omega) + \sigma'_{\rm reg}(\omega,t),$$
$$D(t) = \frac{1}{2Vt_m}\int_0^{t_m} ds\left[\langle\tau\rangle_{t+s} - \int_0^s ds'\chi(t+s,t+s')\right], \quad (9)$$

again for $t_m \to \infty$. Equation (9) is a generalization of the equilibrium expression, $D = (1/2V)[\langle\tau\rangle - \chi'(\omega=0)]$ [20]. In contrast to the sum rule, $D(t)$ following from Eq. (9) is expected to be dominated by $t',t'' >> t$ in Eq. (6). Its dependence on t is revealed if written in the basis of eigenstates $|\phi_m\rangle$ of H_0. In the standard notation for matrix elements $\langle\phi_m|O|\phi_n\rangle = O_{mn}$ and amplitudes $\langle\phi_m|\psi(0)\rangle = a_m$ ($t = 0$ chosen arbitrarily, e.g., when the nonequilibrium state is prepared), we can express $D(t)$ in the eigenbasis, assuming that there are no degeneracies:

$$D(t) = \frac{1}{V}\sum_m |a_m|^2\left[\frac{\tau_{mm}}{2} - \sum_{n\neq m}\frac{|j_{mn}|^2}{(\epsilon_n-\epsilon_m)}\right]$$
$$+ \frac{1}{2V}\sum_{m,n\neq m} a^*_m a_n \frac{j_{mn}(j_{mm}-j_{nn})}{(\epsilon_m-\epsilon_n)}\,e^{i(\epsilon_m-\epsilon_n)t}. \quad (10)$$

As relevant for the isotropic cases $D(t) = D_{\alpha\alpha}(t)$ is the diagonal term, with $j = \mathbf{j}_\alpha$. Obviously, the last term in Eq. (10) provides dependence on t if nonzero. However, it is expected to vanish if averaged over t [15], yielding stationary $D(t) = D_0$ which is dependent only on the (initial) nonequilibrium state $|\psi(0)\rangle$ through a_m. Moreover, the first two terms in Eq. (10) resemble the equilibrium expression [21], with thermal weights substituted by projection weights $|a_m|^2$. However, the latter derivation is feasible only for the time independent H_0. One can express limiting D_0 also for the case with degeneracies, where it is of a more general form:

$$D_0 = \frac{1}{V}\sum_{\epsilon_m=\epsilon_n} a^*_m a_n\left[\frac{\tau_{mn}}{2} - \sum_{\epsilon_l\neq\epsilon_m}\frac{j_{ml}j_{ln}}{(\epsilon_l-\epsilon_m)}\right], \quad (11)$$

still being independent of the choice of the basis within the degenerate sector.

We have so far considered the response of a pure state $|\psi(t)\rangle$ and a t-independent unperturbed H_0. The formalism can be extended also to the more general density matrix as well as the time dependent $H_0(t)$, e.g., representing the presence of the pump. In this case, the response to the perturbation $V(t) = f(t)V$ for an ensemble of pure states or time dependent $H_0(t)$ is derived by expanding the density matrix up to the first order in $f(t)$, $\rho(t) = \rho_0(t) + \rho_1(t) + O(f^2)$, and using the von Neumann equation $\partial\rho(t)/\partial t = -i[H_0(t) + f(t)V,\rho(t)]$. The linear response of general operator B at time t' to perturbation applied at t is then

$$\delta\langle B\rangle_{t'} = \mathrm{Tr}[\rho_1(t')B]$$
$$= -i\int_t^{t'} dt''\,f(t'')\mathrm{Tr}[\rho_0(t)[B^I(t'),V^I(t'')]], \quad (12)$$

$$\rho_0(t) = U(t,0)\rho_0(0)U^\dagger(t,0),$$
$$\rho_1(t') = i\int_t^{t'} dt'' f(t'')U(t',t'')[\rho_0(t''),V]U^\dagger(t',t''), \quad (13)$$

where the time-evolution operator is $U(t',t) = \hat{T}[\exp(-i\int_t^{t'} H_0(t'')dt'')]$. In such a formulation the linear response of the particle current to the field applied at t, written with the density matrix for a single pure state $\rho_0(t) = |\psi(t)\rangle\langle\psi(t)|$ is

$$\delta\langle\mathbf{j}\rangle_{t'} = -i\int_t^{t'} dt''(-e\mathbf{A}(t''))\mathrm{Tr}[\rho_0(t)[\mathbf{j}^I(t'),\mathbf{j}^I(t'')]]. \quad (14)$$

Optical conductivity, possibly generalized also to an ensemble of pure states, is then

$$\sigma(t',t) = \frac{e^2}{V}\left(\langle\tau\rangle_{t'} - i\int_t^{t'} dt''\mathrm{Tr}[\rho_0(t)[\mathbf{j}^I(t'),\mathbf{j}^I(t'')]]\right), \quad (15)$$

where now $\langle\tau\rangle_t = \mathrm{Tr}[\rho_0(t)\tau]$.

III. NUMERICAL IMPLEMENTATION

Let us discuss here the numerical implementation for $\sigma(\omega,t)$ only for the isotropic system in the case of a single w.f. $|\psi(t)\rangle$ and time independent H_0. The apparent disadvantage of the definition with Eq. (6) is that for a fixed t the evaluation of $\chi(t',t'')$ via Eq. (4) requires the propagation $U(t',t'')$ for each t''. Finally, for the numerical calculation at chosen t_m, $\mathcal{O}(t_m^2/\Delta t^2)$ operations are needed, with Δt being the integration time step.

Instead it is more efficient to calculate the integral inside the matrix element, Eq. (4), performing discrete steps:

$$\int_t^{t'} dt''U(t',t'')j|\psi(t'')\rangle \approx \Delta t\sum_{n=0}^{n_m} U_\Delta^{n_m-n+1}\, j|\psi(t+n\Delta t)\rangle, \quad (16)$$

where $n_m = (t'-t)/\Delta t - 1$ and $U_\Delta = U(t''+\Delta t,t'') = \tilde{U}(\Delta t)$ propagation for Δt. The sum Eq. (16) can be then evaluated recursively:

$$|S_0\rangle = j|\psi(t)\rangle,\ |S_n\rangle = j|\psi(t+n\Delta t)\rangle + U_\Delta|S_{n-1}\rangle, \quad (17)$$

so that finally

$$\sigma(t',t) = (e^2/V)(\langle\tau\rangle_{t'} + 2\mathrm{Im}\langle\psi(t')|j\ U(\Delta t)|S_{n_m}\rangle). \quad (18)$$

Such a procedure reduces the number of operations to $\mathcal{O}(t_m/\Delta t)$. We note that quite an analogous procedure can be applied for other transient correlation functions or for the time dependent $H_0(t)$.

IV. SINGLE EXCITED PARTICLE

In order to test the feasibility of the above formalism and contribute to the discussion of transient optical response of nonequilibrium strongly correlated systems, we investigate in the following the case of a single excited charge carrier (hole) $N_h = 1$, doped in an antiferromagnetic (AFM) Mott-Hubbard insulator. We consider the standard single-band t-J model:

$$H_0 = -t_h\sum_{\langle i,j\rangle,s}\tilde{c}_{i,s}^\dagger\tilde{c}_{j,s} + J\sum_{\langle i,j\rangle}\left(\mathbf{S}_i\cdot\mathbf{S}_j - \frac{1}{4}n_in_j\right), \quad (19)$$

where $\tilde{c}_{i,s} = c_{i,s}(1-n_{i,-s})$ are fermion operators, projected onto the space with no double occupancy, describing hopping between the nearest-neighbor sites only. We consider in the concrete example the 2D square lattice, relevant for cuprates, with $J/t_h = 0.4$. Further on we use $t_h = 1$ as the unit of energy, as well of time $t_0 = \hbar/t_h = 1$. Lattice spacing is set at $a_0 = 1$ so that $V = N$, as well as $e = 1$.

The intention is to consider the situation relevant for the pump-probe experiments on cuprates [1,2]. One can imagine two different situations:

(1) First is the photodoping of the Mott-Hubbard insulator, where a low concentration of highly excited charge carriers (holons and doublons) is created within an otherwise insulating AFM system. Both types of carriers would exhibit, at least in the transient stage, an independent response to the probe pulse. To simulate this situation we consider as the initial w.f. a single hole localized on one site, $|\psi(0)\rangle$ being the eigenstate of the model, Eq. (19), where effective t_h is put to zero.

(2) Another setting is a weakly doped AFM insulator, represented by the g.s. of a single hole within the t-J model. The effect of a strong pump pulse applied to it can be simulated by introduction of a phase shift in the hopping term of Eq. (19), changing $t_h \to t_h^{ij} = t_h\exp(i\theta_{ij})$, where we perform the maximum shift in the chosen direction x, i.e., $\theta_x = \pi$. Both scenarios correspond to the same change in the total kinetic energy E_{kin}, calculated from the expectation value of the t_h term in Eq. (19). As obvious for the initially localized hole with $t_h(t=0) = 0$, the excited state has $E_{\mathrm{kin}}(t=0) = 0$. For the choice $\theta_x = \pi$ the effect of the particular phase shift is to change the sign of hopping in the x direction. Due to rotational invariance this again yields zero total kinetic energy.

Results for a single hole within the t-J model are obtained via two numerical methods. One is the exact diagonalization (ED) of small systems employing the Lanczos method, where we study the 2D square lattices with $N \leqslant 26$ sites and PBC. First we find the g.s. $|\psi(0)\rangle$. By solving the time dependent Schrödinger equation, the time evolution of $|\psi(t')\rangle$ and evaluation of recursive relations, Eq. (17), are obtained by employing the Lanczos basis [22,24]. Since the available square lattices ($N = 18,20,26$) are in general not rotationally invariant we perform the averaging of $\sigma' = (\sigma'_{xx} + \sigma'_{yy})/2$ for case 2 together with corresponding pulses $\theta_\alpha = \theta_x,\theta_y$.

Another method to evaluate $\sigma(\omega,t)$ is the diagonalization within the limited functional space (EDLFS) [23,24] The advantage of the EDLFS method in the equilibrium regime follows from a systematical construction of states with distinct configurations of local spin excitations in the proximity of the hole. In this way (in contrast to the ED on a small system) the method in principle deals with an infinite system. In practice the effective size of the system is larger than in ED, but still limited by the number of basis states taken into account. The EDLFS remains efficient even when applied to nonequilibrium systems, as long as the spin disturbance caused by the local quench remains within the confines of generated

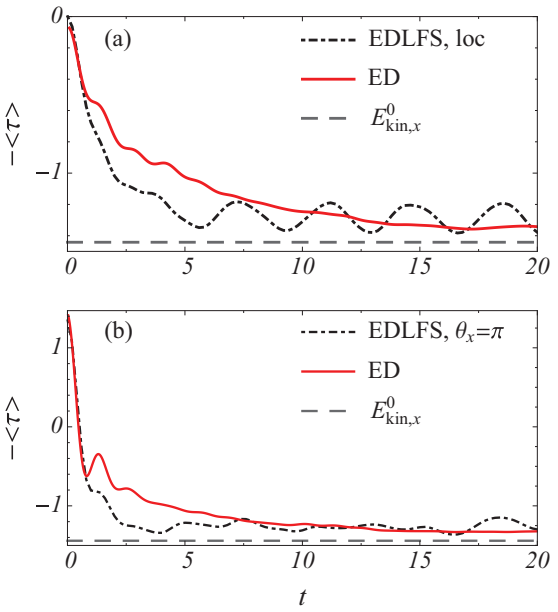

FIG. 1. (Color online) Sum rule $\langle\tau\rangle$ vs time t for $J = 0.4$ as obtained using the ED on $N = 26$ sites as well as the EDLFS. Results are presented for initial states of (a) the localized hole and (b) the π-pulse excited hole. The g.s. $E^0_{\mathrm{kin},x}$ value is also displayed.

spin excitations [25,26]. In the considered case spin excitations extend up to $L = 16$ lattice sites away from the hole.

First we analyze the time variation of $\langle\tau_{\alpha\alpha}\rangle$, representing the sum rule Eq. (8), which is for the tight-binding model Eq. (19) related to the kinetic energy, $\langle\tau_{\alpha\alpha}\rangle_t = -E_{\mathrm{kin},\alpha}(t)$, where only hopping in the α direction is taken into account. In Fig. 1 we present ED and EDLFS results for $\langle\tau\rangle$ calculated directly from kinetic energy for $|\psi(0)\rangle$ corresponding to localized and π-pulse excitations, respectively. Averaging $\langle\tau\rangle = (\langle\tau_{xx}\rangle + \langle\tau_{yy}\rangle)/2$ is employed in ED calculations, whereas for EDLFS $\langle\tau\rangle = \langle\tau_{xx}\rangle$. For comparison we show also the g.s. $E^0_{\mathrm{kin},x} = E^0_{\mathrm{kin}}/2 \sim -1.4$. From Fig. 1 it follows that for both types of initial excited states the decay of $\langle\tau\rangle$ is very fast. The corresponding short time t_d can be related to the formation of the spin polaron and can be explained with the generation of string states [27]. It is expected to scale as $t_d \propto (J/t_h)^{-2/3}$.

We confirm in Fig. 1 that both methods, the ED and the EDLFS, give quite consistent results for $\langle\tau\rangle_t$ for short times $t < t_d \sim 1.5$ (for chosen $J = 0.4$). For intermediate times $t_d < t < t_i \sim 15$ the decay of $E^0_{\mathrm{kin},x}$ evaluated with ED is somewhat slower, which could indicate the influence of finite-size and PBC effects. Namely, in small systems considered with the ED, spin excitations populate the lattice and consequently influence further hole relaxation. The spread of spin excitation is expected to saturate in $t_i \sim \sqrt{N}/J$, yielding an approximately stationary response afterwards. Within the EDLFS with in principle an infinite lattice, such effects are not present or appear only at later times due to the restricted basis. More than an artifact, ED results on small lattices should be relevant for the optical sum rules of systems with finite density of carriers, while those from EDLFS correspond to systems with vanishing density.

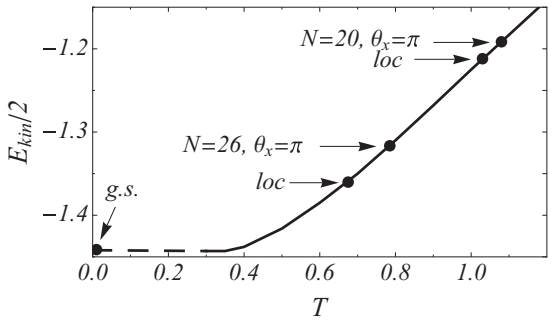

FIG. 2. Equilibrium kinetic energy $E_{\mathrm{kin}}/2$ vs temperature T for a single hole on a system with $N = 26$ sites. Dots mark $E_{\mathrm{kin}}(t \to \infty)/2$ for both types of excitations in $N = 20,26$ systems and g.s.

Although for both types of excitations $E_{\mathrm{kin}}(t = 0) \sim 0$, the distinction in sum rules is apparent for times shorter than $t < t_d$, featuring the fact that the state of the initially localized hole has the rotational symmetry, whereas with the π pulse this symmetry is broken, with $E_{\mathrm{kin},x}(t = 0) = -E_{\mathrm{kin},y}(t = 0)$.

It is evident that for long times $t > t_i$ kinetic energy approaches or oscillates around a quasistationary value, denoted by $E_{\mathrm{kin}}(t \to \infty)$. Within the ED, the latter still somewhat depends on the system size N. The larger the system under consideration, the closer this value is to the g.s. E^0_{kin}, as marked in Fig. 2.

Since the ED simulates a fixed-size system, and the excited state is quite far from the g.s., a plausible interpretation could be investigated within the concept of thermalization, i.e., the approach to the equilibrium state with a finite effective temperature $T_{\mathrm{eff}} > 0$.

One could argue that different $E_{\mathrm{kin}}(t \to \infty)$ originate in different T_{eff}, depending on the type of quench and size of the system. In a finite system T_{eff} is set by the excitation energy so that the canonical expectation value of the energy equals the total initial energy $\langle\psi(0)|H_0|\psi(0)\rangle = E_{\mathrm{tot}}$. At $N = 26$ the effective temperature is approximately $T_{\mathrm{eff}} = 0.35,0.5$ for the initially localized hole and π-pulsed hole, respectively, and increases as N decreases.

Our observation is that $E_{\mathrm{kin}}(T_{\mathrm{eff}})$ is still lower than $E_{\mathrm{kin}}(t \to \infty)$ for all finite systems considered, as seen in Fig. 2. This suggests that the system cannot completely thermalize, possibly due to the discreteness of spin excitations in finite systems. In this connection we notice that the $T > 0$ calculation of the same model, Eq. (19), with a single hole using the finite temperature Lanczos method [24,28] surprisingly reveals that $E_{\mathrm{kin}}(T \sim J)$ does not essentially differ from g.s. E^0_{kin}, Fig. 2.

In Fig. 3 we finally present results for the time dependent optical spectra per hole $\tilde{\sigma}'(\omega,t) = N\sigma'(\omega,t)$ as obtained by ED and EDLFS, respectively, following the described procedure. The cases of initially localized and π-pulse excited holes are compared. With respect to Fig. 1 chosen times represent different evolution stages: (a) $t \sim 0$ response of the initial excited state $|\psi(0)\rangle$, (b) response at approximately characteristic decay time $t = 1.5 \sim t_d$, and (c) $t = 10 \sim t_i$ already relaxed but not yet fully stationary response. To mimic the stationary response for finite systems, $\tilde{\sigma}'(\omega,\bar{t})$, obtained by average over responses in interval $t \in [20,60] \gg t_d$, is presented in Fig. 4 and compared with the g.s. $\tilde{\sigma}_0(\omega)$ and with

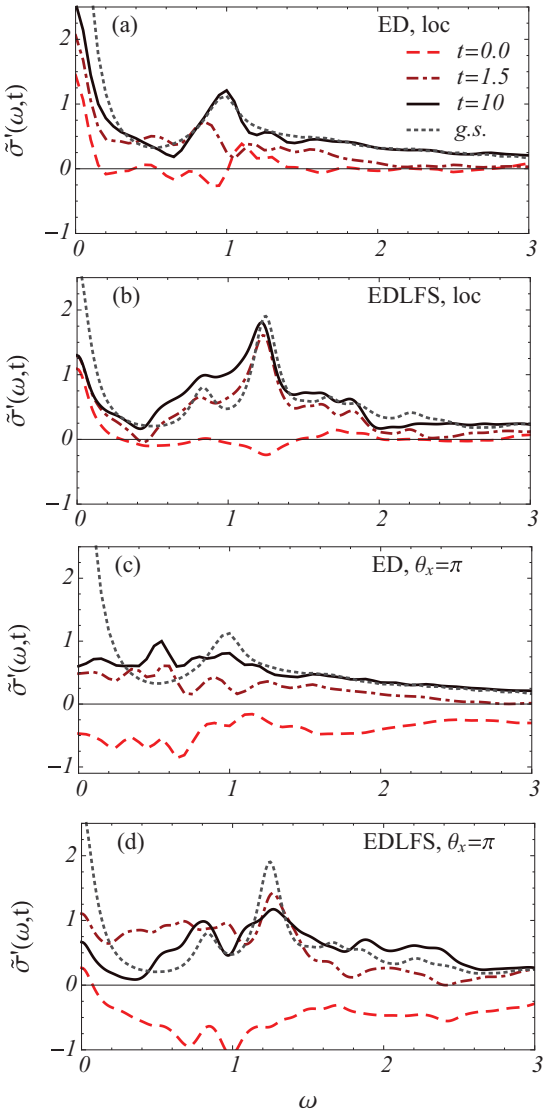

FIG. 3. (Color online) Time dependent optical conductivity $\tilde{\sigma}'(\omega,t)$ (per hole) vs ω as calculated at different times $t = 0,1.5,10$ for (a) the initially localized hole with the ED on $N = 26$ sites, (b) the localized hole with the EDLFS, (c) the π-pulse excited hole with the ED, and (d) the π-pulse excited hole within EDLFS. For comparison the g.s. $\tilde{\sigma}_0(\omega)$ is shown. Broadening of spectra $\delta\omega = 0.1$ is used.

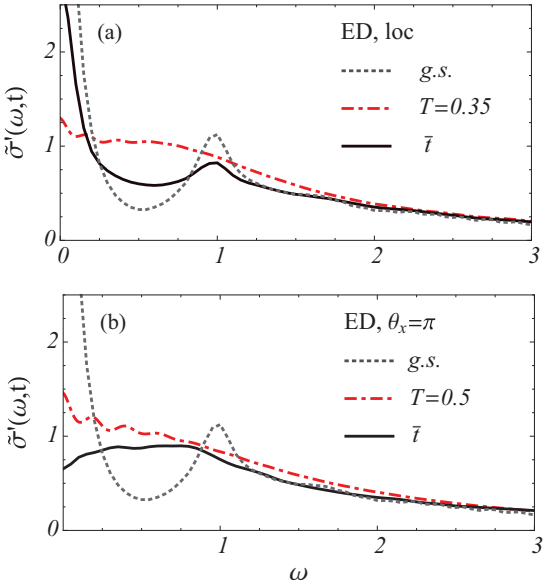

FIG. 4. (Color online) Long-time response $\tilde{\sigma}'(\omega,\bar{t})$ vs ω as calculated with ED on $N = 26$, obtained by average over responses in interval $t \in [20,60] \gg t_d$ for (a) the initially localized hole and (b) the π-pulse excited hole. Results are compared to the g.s. $\tilde{\sigma}_0(\omega)$ and the thermal $\tilde{\sigma}_{\rm th}(\omega)$ at corresponding effective temperatures $T = 0.35,0.5$. Spectra are broadened with $\delta\omega = 0.1$.

the thermal-equilibrium result $\tilde{\sigma}_{\rm th}(\omega)$ at effective $T_{\rm eff} \gtrsim J$, all obtained within the ED.

In the initial stage the response is very incoherent and for the π pulse case even predominantly negative $\tilde{\sigma}'(\omega,t) < 0$, which is compatible with the sum rule $\langle\tau\rangle_{t\sim 0} < 0$ in Fig. 1. The latter indicates a highly nonequilibrium state $|\psi(0)\rangle$ corresponding to an inverse particle population [29].

With some quantitative difference between both methods, within the g.s. $\tilde{\sigma}_0(\omega)$ two features are well visible and of particular interest. One is the midinfrared (MIR) peak at $\omega \sim 2.4J$, which is the signature of the stringlike excited particle states within the 2D AFM background. We see in Figs. 3(a) and 3(b) that the MIR-like peak appears also in $\tilde{\sigma}(\omega,t)$ and stabilizes very fast at $t \sim t_d$ for the localized hole, independent of the numerical method employed. On the other hand, after the π pulse [Figs. 3(c) and 3(d)] the MIR peak is less pronounced. Especially within the ED, $\tilde{\sigma}(\omega,t > t_d)$ is closer to the thermal $\tilde{\sigma}_{\rm th}(\omega)$ at appropriate $T_{\rm eff} = 0.5$. In the equilibrium at such high $T > J$ the MIR peak [Figs. 3(c) and 4(b)] is already smeared out due to thermally disordered AFM spin background. Within the EDLFS results [Fig. 3(d)] the MIR peak recovers better, which could be attributed to a lower density of spin excitations in effectively bigger systems.

The second feature pronounced within the g.s. is the Drude weight D, i.e., peak at $\omega = 0$, which accounts for $\sim 1/3$ of the weight in the sum rule at $T = 0$. It should acquire a finite width in the equilibrium at $T > 0$ due to scattering processes (i.e., in the strict sense $D = 0$ is expected for a nonintegrable system). Although within both methods used [Eq. (10) would require the full ED method] we cannot strictly establish and determine the value of $D(t)$ as defined in Eqs. (9) and (10), it is evident that, with respect to the low-ω response studied, excited particles reveal quite different behaviors. Within the ED the π-pulse excited hole shows essentially no Drude peak, i.e., no corresponding low-ω remainder at $t > 0$. On the other hand, the initially localized hole displays a substantial low-ω peak and presumably $D(t) > 0$ (note that we use broadening $\delta\omega = 0.1$) at all $t > 0$, although the weight is smaller than in $\tilde{\sigma}_0(\omega)$. Using the EDLFS we notice a weak remainder of the low-ω contribution even for the π pulse, though much smaller than for the localized hole. These findings indirectly support Eq. (10), that also the initial w.f. (and not just its total energy) determines the limiting Drude weight value D_0.

V. CONCLUSIONS

We have presented a formalism for the linear optical conductivity response $\sigma(\omega,t)$ of a nonequilibrium (excited) state of a strongly correlated system, where the probe pulse is taken as a perturbation in the linear order. In the absence of a unique approach we have chosen the definition reflecting the onset of the probe electric-field pulse at time t, which is in contrast to some other studies. Such an approach allows the discussion of the optical sum rule as well as the dissipationless Drude weight at any time $t > 0$. On the other hand, the definition introduces a complication due to an additional time integration which we circumvent by a particular numerical implementation.

The presented test case of a single highly excited charge carrier within the t-J model already shows several features and opens questions relevant for the theoretical analysis of the pump-probe spectroscopy results. Independently of the initial state we observe a fast relaxation of several observables, e.g., the kinetic energy and the optical sum rule, toward the respective g.s. value. Still, more specific features of the transient and long-time optical response, such as the MIR peak and the Drude component $D(t)$, appear nonuniversal. Our results reveal that they do not depend merely on the excitation energy, as, e.g., expected from the canonical thermalization, but as well on the character and w.f. of the initial excited state. The persistence of this feature in the thermodynamic limit remains an open question. On one hand, it appears plausible that in an infinite system the local state of the quasiparticle will correspond to the ground state. Nevertheless, this statement is, e.g., not evident for a quantum system with gapped bosonic excitations. To this end our findings show a lack of canonical thermalization, observed before in theoretical [30,31] as well as experimental studies [32]. Since we address excited systems, the concepts of thermalization and relaxation to the g.s. response are only partly applicable, especially for dynamical quantities, and remain the challenge also for further studies. We should note as well that in the application of our formalism and results to the pump-probe experiments some care is needed when energy absorption of particular probe pulses is measured, which cannot be directly compared to time dependent $\sigma'(\omega,t)$ calculated in the present study.

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[1] K. Matsuda, I. Hirabayashi, K. Kawamoto, T. Nabatame, T. Tokizaki, and A. Nakamura, Phys. Rev. B **50**, 4097 (1994).
[2] H. Okamoto, T. Miyagoe, K. Kobayashi, H. Uemura, H. Nishioka, H. Matsuzaki, A. Sawa, and Y. Tokura, Phys. Rev. B **82**, 060513 (2010); **83**, 125102 (2011).
[3] S. Wall *et al.*, Nature Phys. **7**, 114 (2011).
[4] F. Novelli, D. Fausti, J. Reul, F. Cilento, P. H. M. van Loosdrecht, A. A. Nugroho, T. T. M. Palstra, M. Grüninger, and F. Parmigiani, Phys. Rev. B **86**, 165135 (2012).
[5] S. Dal Conte *et al.*, Science **335**, 1600 (2012).
[6] C. Giannetti *et al.*, Nature Communications **2**, 353 (2011).
[7] M. Eckstein and P. Werner, Phys. Rev. B **84**, 035122 (2011).
[8] M. Eckstein and P. Werner, Phys. Rev. Lett. **110**, 126401 (2013).
[9] Z. Lenarčič and P. Prelovšek, Phys. Rev. Lett. **111**, 016401 (2013).
[10] A. Shimizu and T. Yuge, J. Phys. Soc. Jpn. **80**, 093706 (2011).
[11] M. Eckstein and M. Kollar, Phys. Rev. B **78**, 205119 (2008).
[12] M. Eckstein, M. Kollar, and P. Werner, Phys. Rev. B **81**, 115131 (2010).
[13] J. K. Freericks, H. R. Krishnamurthy, and T. Pruschke, Phys. Rev. Lett. **102**, 136401 (2009).
[14] G. De Filippis, V. Cataudella, E. A. Nowadnick, T. P. Devereaux, A. S. Mishchenko, and N. Nagaosa, Phys. Rev. Lett. **109**, 176402 (2012).
[15] D. Rossini, R. Fazio, V. Giovannetti, and A. Silva, arXiv:1310.4757.
[16] L. Foini, L. F. Cugliandolo, and A. Gambassi, Phys. Rev. B **84**, 212404 (2011).
[17] L. Foini, L. F. Cugliandolo, and A. Gambassi, J. Stat. Mech. (2012) P09011.
[18] B. Moritz, A. F. Kemper, M. Sentef, T. P. Devereaux, and J. K. Freericks, Phys. Rev. Lett. **111**, 077401 (2013).
[19] P. Nozières, *Theory of Interacting Fermi Systems* (Addison-Wesley, Reading, 1964).
[20] J. Jaklič and P. Prelovšek, Adv. Phys. **49**, 1 (2000).
[21] H. Castella, X. Zotos, and P. Prelovšek, Phys. Rev. Lett. **74**, 972 (1995).
[22] T. J. Park and J. C. Light, J. Chem. Phys. **85**, 5870 (1986).
[23] J. Bonča, S. Maekawa, and T. Tohyama, Phys. Rev. B **76**, 035121 (2007).
[24] For a review see P. Prelovšek and J. Bonča, in *Strongly Correlated Systems—Numerical Methods*, edited by A. Avella and F. Mancini, Springer Series in Solid-State Sciences Vol. 176 (Springer, Berlin, 2013), pp. 1–29.
[25] M. Mierzejewski, L. Vidmar, J. Bonča, and P. Prelovšek, Phys. Rev. Lett. **106**, 196401 (2011).
[26] L. Vidmar, J. Bonča, T. Tohyama, and S. Maekawa, Phys. Rev. Lett. **107**, 246404 (2011).
[27] D. Golež, J. Bonča, M. Mierzejewski, and L. Vidmar, arXiv:1311.5574.
[28] M. M. Zemljič and P. Prelovšek, Phys. Rev. B **72**, 075108 (2005).
[29] N. Tsuji, T. Oka, and H. Aoki, Phys. Rev. Lett. **103**, 047403 (2009).
[30] M. Rigol, V. Dunjko, and Maxim Olshanii, Nature (London) **452**, 854 (2008).
[31] S. R. Manmana, S. Wessel, R. M. Noack, and A. Muramatsu, Phys. Rev. Lett. **98**, 210405 (2007).
[32] C. Giannetti, G. Zgrablic, C. Consani, A. Crepaldi, D. Nardi, G. Ferrini, G. Dhalenne, A. Revcolevschi, and F. Parmigiani, Phys. Rev. B **80**, 235129 (2009).