

## Photoinduced Drude weights critically enhanced by charge fluctuations in a one-dimensional Mott insulator

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This Letter theoretically validated that the  $\omega = 0$  component of a photoexcited Drude peak in one-dimensional Mott insulators is nonzero only when a finite amount of charge fluctuations is included. Here, we define charge fluctuations as annihilations and creations of photoexcited elementary particles, that are, holons (Hs) and doublons (Ds). In contrast, when the electron on-site repulsion,  $U$ , is infinite, such fluctuations are completely suppressed, and the H and D behave, such as rigid balls, leading to a vanishing  $\omega = 0$  component. Such behaviors inspire us to name them as “quantum clackers.” Moreover, we demonstrated that the effective broadenings of the finite-frequency components of the Drude peak are affected by charge fluctuations and carrier density. Finally, we discovered that such broadenings can be estimated using the group velocity of the relative motion of an H and a D as would be predicted from a colliding picture of the two particles.

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**Introduction.** Recent advancements in ultrafast laser technology for controlling the temporal width, wavelength, and phase of a pulse have facilitated the creation of tunable and novel quantum states for various materials [1–4]. Among such studies, photoinduced insulator-metal transition phenomena in strongly correlated electron systems have been positioned as a fundamental issue that has been attracting extensive attention for a long time [5–18]. Many experimental [19–29] and theoretical [30–45] studies have reported the significance and utility of photoexcited pairs of a holon (H) and a doublon (D) in the descriptions of the low-energy optical excitations. However, these studies were unable to fully clarify the microscopic picture of the electric transport induced by such photoexcited carriers. In this Letter, we theoretically discuss this point based on a one-dimensional (1D) Mott insulator, paying special attention to the motion of the holon-doublon (HD) pair.

In general, a Drude weight, defined as a direct current component of electrical or optical conductivity, is a useful quantity for characterizing metals or insulators [46]. In the case of photoinduced phenomena, we can observe this quantity as the nonzero signal at approximately zero frequency of pump-probe spectra at early timescales. Currently, numerous studies have revealed the features of Drude weights of Mott insulators [47–59]. In addition to these previous studies, herein, we focus on two important factors that determine the Drude weights: carrier density associated with the strength of pumping lights and the degree of charge fluctuations corresponding to the HD pair creations and annihilations.

Starting from a Mott insulator ground state with no HD pair in a 1D  $N$  site ring as shown in Fig. 1(a), we focus on the case of a single photodoped HD pair, which is schematically shown in Fig. 1(c). We have also included the case of a single doped hole for comparison in Fig. 1(b). An induced metallic state is

characterized by a carrier of a single HD pair for the former and a single hole for the latter. Here, our theoretical suggestion is that the metallic feature of the photodoping carriers with finite  $U$  is completely different from that with large  $U$ . Here,  $U$  is the electron mutual repulsion on the same site, namely, the so-called Hubbard  $U$ , which yields a Mott insulator. Another important energy is the degree of electron itineracy for which the electron transfer energy between the nearest-neighboring sites  $T$  is chosen. In the case of  $U \gg T$ , an H and a D seldom exchange their spatial positions as illustrated in Fig. 1(c). For such an exchange, we need an annihilation and a creation of the pair, which leads to the matrix element of the order of  $T^2/U$ . Since the two carriers should move in opposite directions under a uniform electric field, such a forbidden exchange leads to repeated collisions between H and D. This situation corresponds to vanishing Drude weight or DC conductivity. In contrast, in the case of  $U \sim T$ , such restrictions are relaxed in the sense that they penetrate each other owing to the tunneling effect and provide a finite Drude weight, in turn. Such behaviors, namely, completely rigid balls for  $U \gg T$  and partially transparent balls for  $U \sim T$ , are like “quantum clackers.”

To confirm the above hypothesis, we theoretically evaluate the Drude weights of photoexcited states using two effective models based on the charge model [60]. The charge model has been introduced as an effective model of a 1D half-filled extended Hubbard model under the periodic boundary condition and spin-charge separation picture [61–70]. Although this model neglects the spin degrees of freedom, it precisely treats the charge fluctuations, namely, the creations and annihilations of the HD pairs. Consequently, we can reproduce low-energy photoexcited properties of the original Hubbard model [60,71,72]. Another important point is the choice of the system size ( $N$ ). Based on the picture of colliding an H

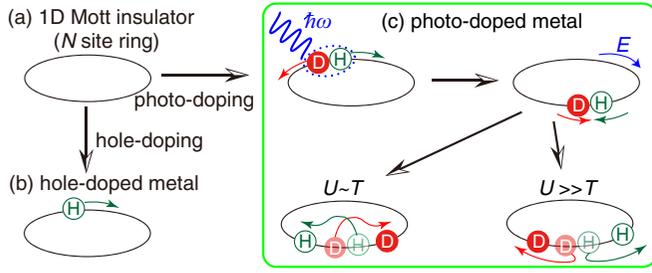


FIG. 1. Schematic of metallic states in a 1D Mott insulator induced by hole doping and photodoping. The wavy line represents irradiated light.  $U$  and  $T$  denote the strength of the on-site Coulomb interaction and transfer energy, respectively. The curved arrows represent the motions of H and D. The electric field of the probe pulse is denoted as  $E$ .

and a D, we also think that the number density of the HD pairs is essential. However, at the present stage, it is technically hard to analyze the effect of the density as we change both the system size and the pair number simultaneously. We, therefore, only change  $N$ , whereas keeping the pair number at one and investigate the dependence on the density that is defined as  $n_c = 1/N$ . Throughout this Letter, we assume the absolute zero temperature and set  $\hbar = e = c = a = 1$ , where  $a$  is a lattice constant.

*Formulations.* We first introduce the charge model defined for a ring of even  $N$  sites under a zero center-of-gravity momentum frame. According to our previous studies [60,71], the model Hamiltonian and charge-current operator  $J$  can be expressed by the field theoretical description of hard-core bosons ( $U_c \rightarrow +\infty$ ) as follows:

$$H \equiv H_T + H_U + H_V + U_c \sum_{j=1}^N (d_j^\dagger d_j^\dagger d_j d_j + h_j^\dagger h_j^\dagger h_j h_j + d_j^\dagger d_j h_j^\dagger h_j), \quad (1)$$

$$H_T \equiv -T \sum_{j=1}^N (d_{j+1}^\dagger d_j + h_{j+1}^\dagger h_j + \text{H.c.}) - \sqrt{2}c_s T \sum_{j=1}^N (d_{j+1}^\dagger h_j^\dagger + h_{j+1}^\dagger d_j^\dagger + \text{H.c.}), \quad (2)$$

$$H_U \equiv \frac{U}{2} \sum_{j=1}^N (n_j^{(d)} + n_j^{(h)})$$

$$H_V \equiv \sum_{\alpha} V_{\alpha} \sum_{j=1}^N (n_{j+\alpha}^{(d)} n_j^{(d)} + n_{j+\alpha}^{(h)} n_j^{(h)} - n_{j+\alpha}^{(d)} n_j^{(h)} - n_{j+\alpha}^{(h)} n_j^{(d)}), \quad (3)$$

$$J \equiv iT \sum_{j=1}^N (d_{j+1}^\dagger d_j - h_{j+1}^\dagger h_j - \text{H.c.}) + i\sqrt{2}c_s T \sum_{j=1}^N (d_{j+1}^\dagger h_j^\dagger - h_{j+1}^\dagger d_j^\dagger - \text{H.c.}). \quad (4)$$

Here,  $h_j$  ( $h_j^\dagger$ ) and  $d_j$  ( $d_j^\dagger$ ) are the annihilation (creation) operators at the  $j$ th site of an H and a D, respectively.  $n_j^{(h)} = h_j^\dagger h_j$ ,  $n_j^{(d)} = d_j^\dagger d_j$ , and  $h_{N+1}^{(\dagger)} = h_1^{(\dagger)}$  ( $d_{N+1}^{(\dagger)} = d_1^{(\dagger)}$ ).  $V_{\alpha} \equiv V/\alpha$  is the  $\alpha$ th nearest-neighbor Coulombic energy. In this Letter, we set  $c_s = 0.82$ , based on our previous works [60,71,72].  $c_s$  is a kind of correction factor, which is related to the spin degrees of freedom of an original 1D half-filled Hubbard model. The  $U_c$  term is equivalent to  $d_j^2 = h_j^2 = (d_j^\dagger)^2 = (h_j^\dagger)^2 = d_j h_j = 0$ , and conventional bosonic commutation relations,  $[d_j, d_k^\dagger] = [h_j, h_k^\dagger] = \delta_{j,k}$ ,  $[d_j, d_k] = [h_j, h_k] = [d_j, h_k] = 0$ , and  $[d_j^\dagger, d_k^\dagger] = [h_j^\dagger, h_k^\dagger] = [d_j^\dagger, h_k^\dagger] = 0$  hold. Note that we need not explicitly treat the  $U_c$  term in actual calculations when we prepare basis states appropriately.

Defining an operator of translating one site to the right as  $T_R$ , parity inversion as  $\mathcal{P}$ , and charge conjugation as  $\mathcal{C}$ , we first introduce a symmetrized bare basis,

$$|r_M^\pm\rangle \equiv \frac{1}{\sqrt{N}} \sum_{l=0}^{N-1} T_R^l (1 \pm \mathcal{P})(1 \pm \mathcal{C}) \prod_{p_i \neq q_j, i, j=1}^M d_{p_i}^\dagger h_{q_j}^\dagger |0\rangle, \quad (5)$$

where the sign selections on the right-hand side are limited to  $(+, +)$  and  $(-, -)$  for  $|r_M^+\rangle$  and  $|r_M^-\rangle$ , respectively, and  $M$  denotes the number of HD pairs. Here,  $p_i$  and  $q_j$  are the  $i$ th and  $j$ th positions of D and H, respectively. The Mott-insulator ground state belongs to the  $(+, +)$  subspace, whereas the photoexcited state starting from it belongs to the  $(-, -)$  subspace owing to  $\mathcal{P}^\dagger J \mathcal{P} = \mathcal{C}^\dagger J \mathcal{C} = -J$  and  $\mathcal{P}^\dagger H \mathcal{P} = \mathcal{C}^\dagger H \mathcal{C} = H$ . The projection operator with a fixed  $M$  subspace is defined as

$$P_M^\lambda \equiv \sum_{r_M^\lambda} |r_M^\lambda\rangle \langle r_M^\lambda| \quad (\lambda = \pm). \quad (6)$$

Next, we define our Hamiltonian using this basis set. In this Letter, we select the  $M = 1$  subspace (the subspace of 1-HD-pair states) and prepare two Hamiltonians. The first Hamiltonian is that of the pure HD model without charge fluctuations [32,73], namely,  $H$  in the absence of the second term of Eq. (2), whereas the second Hamiltonian is that of the extended HD model with charge fluctuations. Regarding the latter model, we explain its details later. After solving a Hamiltonian in each case, we obtain corresponding eigenenergies and eigenfunctions,  $E_\mu^\pm$  and  $|\Phi_\mu^\pm\rangle$ , respectively. Here,  $\mu$  takes a number from 1 to  $(N/2 - 1)$  for  $|\Phi_\mu^- \rangle$  and from 1 to  $(N/2)$  for  $|\Phi_\mu^+ \rangle$ , and is related to the relative wave number of the DH motion  $k_\mu$  which is roughly estimated as  $k_\mu = 2\pi\mu/N$ . We can then evaluate the optical conductivity spectrum in a low-energy region with artificial broadening  $\gamma$  and  $\omega_{\mu'\mu} \equiv E_{\mu'}^+ - E_\mu^-$ ,

$$\sigma_\mu(\omega) = \frac{D_\mu}{\pi} \frac{\gamma}{\omega^2 + \gamma^2} + \sum_{\mu'=1}^{N/2} \frac{\gamma}{N\omega_{\mu'\mu}} \left( \frac{|\langle \Phi_{\mu'}^+ | J | \Phi_\mu^- \rangle|^2}{(\omega - \omega_{\mu'\mu})^2 + \gamma^2} + \frac{|\langle \Phi_{\mu'}^+ | J | \Phi_\mu^- \rangle|^2}{(\omega + \omega_{\mu'\mu})^2 + \gamma^2} \right). \quad (7)$$

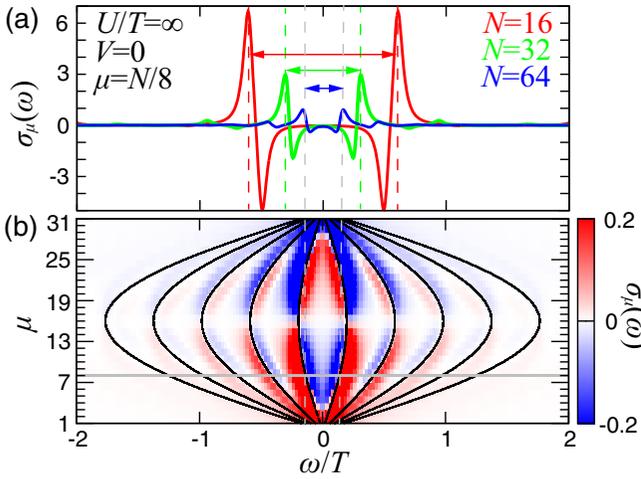


FIG. 2. Optical conductivity spectra of the photoexcited states of the pure HD model for  $V = 0$  and  $\gamma = 0.05T$ . (a)  $\sigma_\mu(\omega)$  at  $\mu = N/8$  for the system size of  $N = 16$  (red),  $N = 32$  (green), and  $N = 64$  (blue). (b)  $\sigma_\mu(\omega)$  at  $N = 64$  for  $1 \leq \mu \leq N/2 - 1$ . The gray horizontal line corresponds to the same spectrum with  $N = 64$  in (a). The black lines in (b) exhibit  $\omega = \pm(2m - 1)(\pi/N)v_g^-(k_\mu)$  for  $m = 1-5$ .

In the first term,  $D_\mu$  is the so-called Drude weight, defined as

$$D_\mu \equiv -\frac{\pi}{N} \langle \Phi_\mu^- | H_T | \Phi_\mu^- \rangle - \frac{2\pi}{N} \sum_{\mu'=1}^{N/2} \frac{|\langle \Phi_{\mu'}^+ | J | \Phi_\mu^- \rangle|^2}{\omega_{\mu'\mu}}, \quad (8)$$

and the second term corresponds to the finite-frequency part of the low-energy excitations. From here on, we refer to the first and second terms as the zero-frequency Drude peak (ZFDP) and the finite-frequency Drude peak (FFDP), respectively. The above formulations in Eqs. (7) and (8) can be derived within the framework of the linear-response theory when one assumes the initial equilibrium state as a certain photoexcited state, which corresponds to  $|\Phi_\mu^- \rangle$  [53]. Here, we have two comments on Eqs. (7) and (8). First, in the summation with respect to  $\mu'$  in the second term of each of them, the terms with  $\omega_{\mu'\mu} = 0$  are excluded by definition. This goes back to their derivations based on the linear-response theory. We can, consequently, perform the summations without any divergence. Second, because of the restriction of the calculation subspace, a type of conservation law, the  $f$ -sum rule, should be satisfied as

$$K_\mu \equiv \lim_{\gamma \rightarrow 0^+} \int_{-\infty}^{\infty} d\omega \sigma_\mu(\omega) = -\frac{\pi}{N} \langle \Phi_\mu^- | H_T | \Phi_\mu^- \rangle. \quad (9)$$

*Pure HD model.* First, we investigate the Drude weights in the pure HD model. In this model, we restrict the subspace to that of  $M = 1$ , switching off the charge fluctuations, which is justified for  $U/T = \infty$ . In more detail, the term of HD annihilation and creation in the Hamiltonian, namely, the second term of Eq. (2) is set to zero.

In the case of  $V = 0$ , we can obtain the analytic solutions for eigenenergies and eigenfunctions [72]. The resulting spectra  $\sigma_\mu(\omega)$  are shown in Fig. 2(a) for  $\mu = N/8$  as an example. In the pure HD model, the eigenenergy is derived as  $E_\mu^- = U - 4T \cos(k_\mu)$  from Eqs. (1) and (2). Since the

above-mentioned  $k_\mu$  is  $\pi/4$ , this is located at the first quarter of the band [see Fig. SM4(a) in the Supplemental Material (SM) [74]]. From the  $f$ -sum rule in Eq. (9),  $K_\mu = (4\pi T/N) \cos(2\pi\mu/N)$  can be derived, the signature of  $\sigma_\mu(\omega)$  being both positive and negative. In the figure, the FFDPs can be clearly seen, whereas the ZFDPs of the photoexcited states do not appear. In fact, from the numerical evaluation of the Drude weight in Eq. (8), we find that  $D_\mu$  vanishes for every  $\mu$  at machine precision as shown in Fig. SM3 of the Supplemental Material ( $U/T = \infty$ ) [74], which indicates that the collision between an H and a D is essential in the understanding of the Drude peak [75].

Concerning the FFDPs, the spectral peaks and dips rigorously appear at  $\omega = \omega_{\mu'\mu}$  for  $\gamma \rightarrow +0$ . The meaning of the overall spectral shape is described in Sec. I of the SM [74]. Substituting  $\mu' = \mu, \mu \pm 1, \mu \pm 2, \dots$  into  $\omega_{\mu'\mu}$ ,  $\omega_{\mu'\mu} = \pm(2m - 1)(\pi/N)v_g^-(k_\mu)$  for  $m = 1, 2, \dots$  is satisfied in the case of  $N \gg 1$  (see Eq. (SM19) in the SM [74]). Here,  $v_g^-(k_\mu) \equiv \partial \varepsilon_\mu^- / \partial k_\mu$  with  $k_\mu = 2\pi\mu/N$  denotes the group velocity of a relative motion of an odd-parity single HD pair. These frequencies coincide with the positions of the aforementioned dips and peaks as shown in Fig. 2(b), although the formers are approximations for an infinite  $N$  and only give averaged values for each pair of a dip and a peak. Since the mean free path of the DH pair equals  $N/2$ , we can estimate  $\tau^* \sim (N/2)/|v_g^-(k_\mu)|$  as an effective lifetime for the DH collision or scattering. The effective broadening defined as  $\pi/\tau^*$  is, consequently, evaluated as  $2\pi|v_g^-(k_\mu)|/N$ , which are specified as the lengths of the horizontal arrows in Fig. 2(a). As seen in the figure, we can confirm coincidences between the estimated effective broadenings and the peak positions. We expect that a Drude peak width will be well defined for a fixed pair-number density  $n_c$  in the limit of infinite  $N$ . Although it is still difficult to determine it in the present calculation, this result suggests that the broadening of the Drude peak is governed by the collision of an H and a D.

For comparison, we briefly comment here on hole-doped 1D systems at a low-density limit. An effective model with  $N$  sites can be obtained by the Hamiltonian  $\tilde{H} = -T \sum_{j=1}^N (h_{j+1}^\dagger h_j + h_j^\dagger h_{j+1})$  and charge-current operator  $\tilde{J} = -iT \sum_{j=1}^N (h_{j+1}^\dagger h_j - h_j^\dagger h_{j+1})$ . First, in the case of a single holon, the eigenmodes are  $|\mu\rangle = (1/\sqrt{N}) \sum_{j=1}^N \exp(i2\pi\mu j/N) h_j^\dagger |0\rangle$  and  $E_\mu = -2T \cos(2\pi\mu/N)$  for  $-N/2 + 1 \leq \mu \leq N/2$ . Using  $|\mu\rangle$  as  $|\Phi_\mu^- \rangle$  in Eq. (8), the photoexcited ZFDP is derived as  $\tilde{D}_\mu = -\pi \langle \mu | \tilde{H}_T | \mu \rangle / N = (2\pi T/N) \cos(2\pi\mu/N)$  [76]. This obviously finite  $\tilde{D}_\mu$  at finite sizes is a striking difference from the above vanishing  $D_\mu$ . Furthermore, we can derive the same quantity for the case of two holons. We select the eigenmode as  $|v\rangle = 1/\sqrt{N} \sum_{i,l} h_{l+i}^\dagger h_l^\dagger f_v(i) |0\rangle$ , where  $l = 1 \sim N$ ,  $i = 1 \sim (N-1)$ , and  $f_v(i) = \sqrt{2/N} \sin(\pi v i/N)$ . Using this selection, we find  $\tilde{D}_v = (4\pi T/N) \cos(\pi v/N)$ , which does not vanish for general  $v$ . This property is intuitively understood when we recall that in this case, all the carriers move in the same direction without collision.

*Extended HD model.* We subsequently investigate the extended HD model to incorporate a finite amount of charge fluctuations. This model is derived using a third-order perturbation of a charge model based on the Schrieffer-Wolff

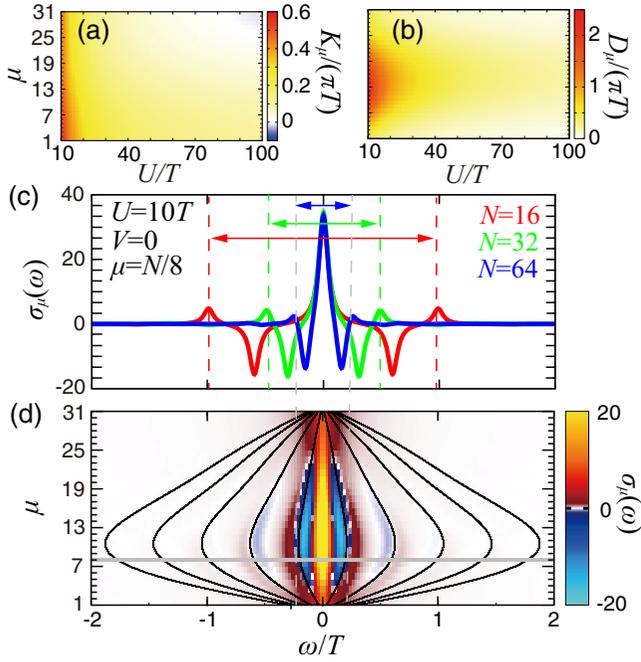


FIG. 3. Optical properties of the third-order perturbative effective model (extended HD model) obtained from a charge model. (a)  $U$  dependency of  $K_\mu$  with  $V = 0$  at  $N = 64$ . (b)  $U$  dependency of  $D_\mu$  with  $V = 0$  at  $N = 64$ . (c)  $\sigma_\mu(\omega)$  for  $U = 10T$ ,  $V = 0$ , and  $\gamma = 0.05T$  at  $\mu = N/8$ . The colors are corresponding to the system size of  $N = 16$  (red),  $N = 32$  (green), and  $N = 64$  (blue). The whole  $\mu$  dependency is shown in (d) for  $N = 64$ . The gray horizontal line corresponds to the same spectrum with  $N = 64$  in (b). The black lines exhibit  $\omega = \pm(2m - 1)(\pi/N)v_g^-(k_\mu)$  for  $m = 1-5$ .

transformation method [77–82]. The quantitative reliability of this method is discussed in Sec. III of the SM [74]. To compare the results with those without fluctuations, we again focus on the photoexcited states associated with the  $M = 1$  subspace in the absence of  $V$ . In this case, all the quantities of  $\omega_{\mu'\mu} \equiv E_{\mu'}^+ - E_\mu^-$ ,  $\langle \Phi_\mu^- | H_T | \Phi_\mu^- \rangle$ , and  $\langle \Phi_\mu^+ | J | \Phi_\mu^- \rangle$  in Eqs. (7)–(9) are automatically corrected in the order of  $1/U^2$  [82]. This means that we can tune the degrees of charge fluctuations by varying the  $U$  value.

Consequently, the key results, including the charge fluctuations with  $V = 0$  are summarized in Figs. 3(a)–3(d). There are two significant differences from the preceding results due to the finite  $U$ . First,  $K_\mu$  determined by the  $f$ -sum rule is positive for  $U \lesssim 20T$  as shown in Fig. 3(a). Second,  $D_\mu$  is positive and finite as seen in Fig. 3(b) and SM3 [74]. For example, even the largest  $U$  case, namely, the case of  $U = 100T$ , also provides finite  $D_\mu$ . Since smaller  $U$  values provide stronger charge fluctuations, the result in Fig. 3(b) and SM3 indicates that strong charge fluctuations significantly enhance the ZFDPs of the photoexcited states. This enhancement is clearly seen in the spectra in Figs. 3(c) and 3(d). We can also estimate the effective broadenings of FFDPs utilizing the same method used for the previous  $\tau^*$  and confirm that the estimated broadenings reproduce the intervals between the peaks quantitatively as shown in Fig. 3(c). We emphasize that the group velocity used here is strongly modified from that without charge fluctuations

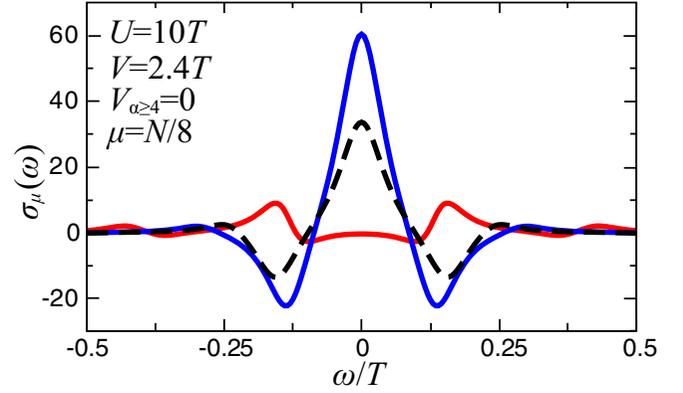


FIG. 4.  $\sigma_\mu(\omega)$  calculated by the extended HD model with  $U = 10T$ ,  $V = 2.4T$ ,  $V_{\alpha \geq 4} = 0$ ,  $N = 64$ , and  $\gamma = 0.05T$  at  $\mu = N/8$  (blue line). The red line is the corresponding result obtained for the pure HD model. The black dashed line for  $V = 0$ , which is the same as the blue line in Fig. 3(c), is shown for comparison.

as shown in Sec. II of the SM [74]. In this sense, we can state that the FFDP is also affected by the charge fluctuations.

Finally, we briefly comment on the results of the finite  $V$ . As is already known, a long-range Coulomb interaction  $V$  ( $V > 0$ ) induces an attractive interaction between an H and a D and makes an HD bound state when the attraction is sufficiently large [29,72]. Meanwhile, the whole spectral shape changes drastically. It is, therefore, worth investigating to confirm whether the aforementioned findings obtained for  $V = 0$  also explain the case of finite  $V$ . In our numerical calculations, the resulting features of  $K_\mu$ ,  $D_\mu$ , and  $\sigma_\mu(\omega)$  for the finite  $V$  are almost qualitatively the same as those for  $V = 0$ . We show one typical example of  $V = 2.4T$  and  $V_{\alpha \geq 4} = 0$  with  $U = 10T$  in Fig. 4. Note that  $\mu$  is  $N/8$ , which means that the initial state is located within the continuum of a free HD pair. Here, the blue (red) line shows the spectrum from the extended (pure) HD model. As an overall feature, the spectrum from the extended HD model shows qualitatively similar features to the corresponding one with  $V = 0$  (dashed black line). In contrast, we note that the weight at  $\omega = 0$  is enhanced from that for  $V = 0$ , which is interpreted as an increased HD exchange rate due to the reduction of the gap energy caused by finite  $V$ . Since this parameter is the best set in our previous work of reproducing the optical spectra of a typical 1D Mott insulator ET-F<sub>2</sub>TCNQ [72], the present result provides an interpretation of future pump-probe measurements for this material.

*Summary.* The nature of photoexcited Drude weights in the 1D Mott insulators was theoretically evaluated using the two effective models, both of which are established based on the spin-charge separation picture. In terms of the linear-response spectra with a certain initial photoexcited state  $\sigma_\mu(\omega)$ , we estimated the properties of both the ZFDPs and the FFDPs associated with a single photogenerated HD pair. Consequently, the former is only finite and enhanced in the presence of charge fluctuations. We also estimated the typical broadenings using the group velocity of the relative motion of a single HD pair for the latter.

As a next step toward the complete physical understanding of the photoinduced metallic state observed in the pump-

probe spectra of Mott insulators, we will evaluate other purely electronic many-body effects, such as the effects of spin degrees of freedom and strong excitations (nonlinear responses), in the near future. Such further exploration will deepen our understanding of the complicated many-body metallic states appearing in the pump-probe spectra of a 1D Mott insulator.

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- [74] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.106.L081119> for (I) the meaning of the spectral shape of the regular part of the optical conductivity defined with a certain excited state as the initial state, (II) the details of the perturbative method and the formulations of the pure HD model and the extended HD model based on it, and (III) the validity and accuracy of the resultant optical properties.
- [75] We also numerically checked that  $D_\mu$  values vanish on the  $M = 2$  subspace with  $V = 0$ . Namely, defining  $P_2^\lambda H P_2^\lambda |\psi_{\mu^\lambda}^\lambda\rangle \equiv \mathcal{E}_{\mu^\lambda}^\lambda |\psi_{\mu^\lambda}^\lambda\rangle$ ,  $D_\mu$  can be calculated by replacing  $\langle \Phi_\mu^- | H_T | \Phi_\mu^- \rangle$ ,  $\langle \Phi_{\mu'}^+ | J | \Phi_\mu^- \rangle$ , and  $\omega_{\mu'\mu}$  with  $\langle \psi_\mu^+ | P_2^+ H_T P_2^+ | \psi_\mu^+ \rangle$ ,  $\langle \psi_{\mu'}^- | P_2^- J P_2^+ | \psi_\mu^+ \rangle$ , and  $\mathcal{E}_{\mu'}^+ - \mathcal{E}_\mu^-$ , respectively.
- [76] In this derivation, the part of the summation does not contribute because the equation of  $\tilde{J}|\mu\rangle = 2T \sin(2\pi\mu/N)|\mu\rangle$  indicates that only the term of  $\mu' = \mu$  survives and that this is excluded in the summation because  $E_{\mu'} = E_\mu$ .
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*Correction:* Two occurrences of the letter  $T$  in the sixth paragraph below Eq. (9) were set incorrectly during the proof production cycle and have been fixed.