

Glassy dynamics of the one-dimensional Mott insulator excited by a strong terahertz pulse

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The elucidation of nonequilibrium states in strongly correlated systems holds the key to emergence of novel quantum phases. The nonequilibrium-induced insulator-to-metal transition is particularly interesting since it reflects the fundamental nature of competition between itinerancy and localization of the charge degrees of freedom. We investigate pulse-excited insulator-to-metal transition of the half-filled one-dimensional extended Hubbard model. Calculating the time-dependent optical conductivity with the time-dependent density-matrix renormalization group, we find that strong mono- and half-cycle pulses inducing quantum tunneling strongly suppress spectral weights contributing to the Drude weight σ_D , even if we introduce a large number of carriers Δn_d . This is in contrast to a metallic behavior of $\sigma_D \propto \Delta n_d$ induced by photon absorption and chemical doping. The strong suppression of σ_D in quantum tunneling is a result of the emergence of the Hilbert-space fragmentation, which makes pulse-excited states glassy.

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Introduction. The elucidation of nonequilibrium states in strongly correlated systems is of great interest since it promises to open a door to the emergence of novel quantum phases. Nonequilibrium quantum many-body states have recently been investigated not only in solids with light and electric fields [1–12] but also in trapped ions [13,14], cold atoms [15–17], and quantum circuits [18–23]. One of the most significant challenges in this field is how to preserve nonequilibrium states, such as the Floquet states [24–26], from thermalization [27–30], for which the realization of many-body localization (MBL) [31–33] may hold the key. Also, the nonequilibrium-induced insulator-to-metal transition is a fundamental issue associated with competition between itinerancy and localization of charge degrees of freedom. The photoinduced insulator-to-metal transitions [2,3,5–8] due to photon absorption have been suggested in the one-dimensional (1D) Mott insulator. Similarly, nonabsorbable terahertz photons with strong intensity have been suggested to induce a metallic state [10,11] via quantum tunneling [34–39].

Until now it has been commonly accepted that the breakdown of the Mott insulators via electric pulses leads to metallic states. However, we raise question about the validity of this understanding. To answer this question, we examine the possibility of the emergence of novel quantum phases such as glass phases with intermediate properties between itinerancy and MBL.

In this Letter, we investigate pulse-excited states of the half-filled 1D extended Hubbard model (1DEHM) using the time-dependent density-matrix renormalization group (tDMRG) [40–42]. We propose a Mott transition to glassy states induced by mono- and half-cycle terahertz pulses. If we excite the Mott insulating state via photon absorption, we obtain metallic states with large spectral weights contributing to the Drude component σ_D . In contrast, we find that strong electric fields inducing the Zener breakdown [43] strongly suppress σ_D , even if we introduce a large number of carriers. We consider that the emergence of the Hilbert-space fragmentation [44–53] due to high fields leads to glassy dynamics [54–59] as seen in fracton systems [60–67].

Model and method. To investigate nonequilibrium properties of the 1D Mott insulator, we use 1DEHM with a vector potential $A(t)$ defined as

$$\mathcal{H} = -t_h \sum_{i,\sigma} B_{i,\sigma} + U \sum_i n_{i,\uparrow} n_{i,\downarrow} + V \sum_i n_i n_{i+1}, \quad (1)$$

where $B_{i,\sigma} = e^{iA(t)} c_{i,\sigma}^\dagger c_{i+1,\sigma} + \text{H.c.}$, $c_{i,\sigma}^\dagger$ is the creation operator of an electron with spin $\sigma (= \uparrow, \downarrow)$ at site i , and $n_i = \sum_\sigma n_{i,\sigma}$ with $n_{i,\sigma} = c_{i,\sigma}^\dagger c_{i,\sigma}$. We consider $(U, V) = (10, 3)$ taking the nearest-neighbor hopping t_h to be the unit of energy ($t_h = 1$), which describes the optical conductivity in a 1D Mott insulator ET-F₂TCNQ [68]. Spatially homogeneous electric field $E(t) = -\partial_t A(t)$ applied along the chain is incorporated via the Peierls substitution in the hopping terms [69]. Unless otherwise noted, we consider the half-filled 1DEHM with $L = 32$ sites. Note that we set the light velocity c , the elementary charge e , the Dirac constant \hbar , and the lattice constant to 1.

We assume that pulses have time dependence determined by $A(t) = A_{\text{pump}}(t) + A_{\text{probe}}(t)$ with $A_{\text{probe}}(t) = A_0^{\text{pr}} e^{-(t-t_0^{\text{pr}})^2 / [2(t_d^{\text{pr}})^2]} \cos[\Omega^{\text{pr}}(t - t_0^{\text{pr}})]$ for probe pulses. Unless

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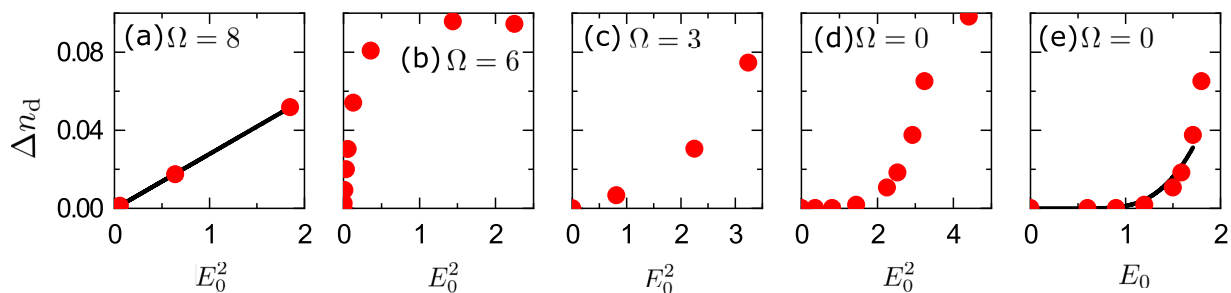


FIG. 1. Δn_d of the $L = 32$ half-filled 1DEHM for $(U, V) = (10, 3)$ excited by electric pulses. Red points are Δn_d as a function of E_0^2 for (a) $\Omega = 8$ with a black line for eye guide, (b) $\Omega = 6$, (c) $\Omega = 3$, and (d) $\Omega = 0$. (e) Red points are Δn_d as a function of E_0 for $\Omega = 0$. The black line shows a fitted curve proportional to $E_0 \exp(-\pi E_{\text{th}}/E_0)$.

otherwise noted, we use $A_{\text{pump}}(t) = A_0 e^{-(t-t_0)^2/(2t_d^2)} \cos[\Omega(t-t_0)]$ for pump pulses. We set $A_0^{\text{pr}} = 0.001$, $\Omega^{\text{pr}} = 10$, $t_d^{\text{pr}} = 0.02$, and $t_0^{\text{pr}} = t_0 + \tau$, where τ indicates the delay time between pump and probe pulses. We obtain time-dependent wave functions by the tDMRG implemented by the Legendre polynomial [70,71] employing open boundary conditions and keep $\chi = 3000$ density-matrix eigenstates. We obtain both singular and regular parts of the optical conductivity in nonequilibrium $\sigma(\omega, \tau) = \frac{j_{\text{probe}}(\omega, \tau)}{i(\omega + i\gamma)L A_{\text{probe}}(\omega)}$ [72–74], where $A_{\text{probe}}(\omega)$ and $j_{\text{probe}}(\omega, \tau)$ are the Fourier transform of $A_{\text{probe}}(t)$ and current induced by a probe pulse, respectively (see Sec. S1 in the Supplemental Material [75]). γ indicates a broadening factor.

Doublon density. First of all, we demonstrate how pumping energy makes a difference in carrier production. Figure 1 shows how much electric pulses with $(t_d, t_0) = (2, 10)$ change doublon density $\Delta n_d = \frac{1}{L}[\langle I \rangle_t - \langle I \rangle_0]$ in 1DEHM, where $I = \sum_j n_{j,\uparrow} n_{j,\downarrow}$, $\langle \mathcal{O} \rangle_t$ is the average of an expectation value of an operator \mathcal{O} from $t = 21$ to 22 just before a probe pulse is applied, and $\langle \mathcal{O} \rangle_0$ is an expectation value of \mathcal{O} for a ground state. We focus on $\Delta n_d < 0.1$, which can be achieved with experiments. Δn_d oscillates even after pulse decay, but their amplitudes are smaller than the radius of red points in Fig. 1. Since $\text{Re}[\sigma(\omega, \tau < 0)]$ has an excitonic level at $\omega = \omega_1$ and a continuum begins at $\omega = \omega_c$ [68,84–87], where $(\omega_1, \omega_c) = (6, 6.5)$ for $(U, V) = (10, 3)$, a pump pulse with $\Omega = 8$ excite electrons in a continuum leading to $\Delta n_d \propto E_0^2$ [see Fig. 1(a)] as discussed in Ref. [39] with the amplitude of electric fields E_0 . Taking $\Omega = \omega_1$, we can efficiently excite doublons and holons even for small E_0 [see Fig. 1(b)]. For subgap excitations, i.e., $\Omega < \omega_1$, electrons are excited by a nonlinear process, which is classified into multiphoton absorption and quantum tunneling. The crossover between them is called the Keldysh crossover [88]. Figures 1(c) and 1(d) show Δn_d generated by two-photon absorption and quantum tunneling, respectively. For $\Omega = 0$ mono-cycle pulses, we find that Δn_d follows a threshold behavior $\Delta n_d \propto E_0 \exp(-\pi E_{\text{th}}/E_0)$ [39] as indicated by the black line in Fig. 1(e). Using this relation, we can estimate the doublon-holon correlation length $\xi \simeq \omega_1/(2E_{\text{th}}) \sim 1.5$.

Glassy dynamics. We show in Fig. 2 the results of 1DEHM excited by a quantum tunneling with strong $\Omega = 0$ pulses whose energy is in terahertz band. We show $\text{Re}[\sigma(\omega, \tau)]$ excited by mono-cycle pulses with $(\Omega, t_d) = (0, 2)$ for

various E_0 in Figs. 2(a)–2(c). $|E(\omega)| = |\int dt e^{i\omega t} E(t)|$ with $E_0 = 1.5$ shown in Fig. 2(d) indicates that the photon energy is too small to excite the Mott gap. We obtain $\Delta n_d = 0.01$, 0.07, and 0.1 for Figs. 2(a)–2(c), respectively. The spectral weights above the Mott gap transfer to lower energies, but we find that the Drude weight σ_D , which we define as spectral weight below $\omega = 0.15$ (see Secs. S2 and S3 in the Supplemental Material [75]), is not proportional to Δn_d but is strongly suppressed even if we take large Δn_d as shown in Figs. 2(b) and 2(c). Note that the Drude weight appears at

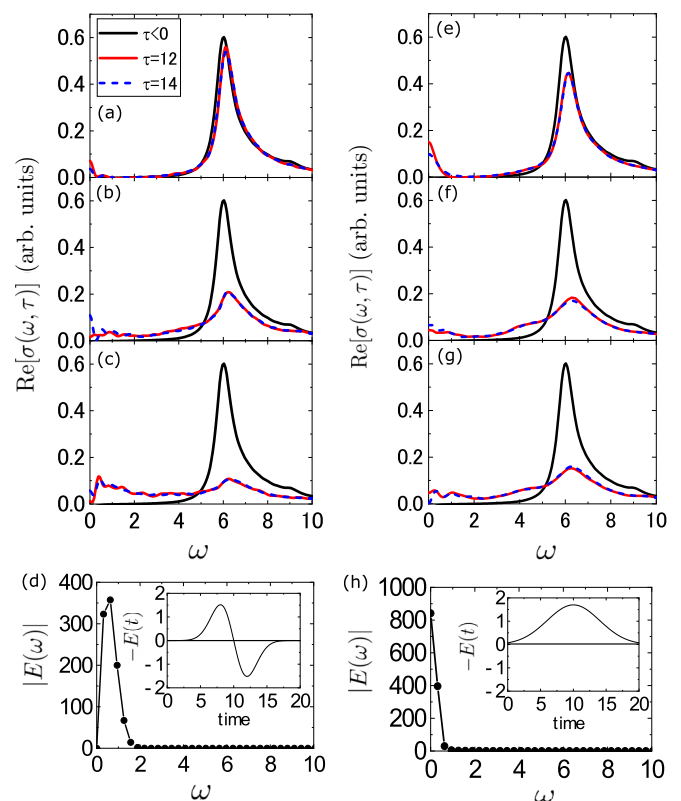


FIG. 2. $\text{Re}[\sigma(\omega, \tau)]$ excited by $\Omega = 0$ mono- [half]-cycle pulses for $t_d = 2$ [$t_d = 4$] with (a) $E_0 = 1.5$, (b) $E_0 = 1.8$, and (c) $E_0 = 2.1$ [(e) $E_0 = 1.7$, (f) $E_0 = 1.9$, and (g) $E_0 = 2.0$]. Black, red, and blue-dashed lines are for $\tau < 0$, $\tau = 12$, and 14, respectively. (d) [(h)] $|E(\omega)|$ of a mono- [half]-cycle pulse with $E_0 = 1.5$ [$E_0 = 1.7$]. The inset indicates $-E(t)$. [(a)–(c)] and [(e)–(g)] are obtained with the half-filled $L = 32$ 1DEHM for $(U, V) = (10, 3)$ taking $\gamma = 0.4$.

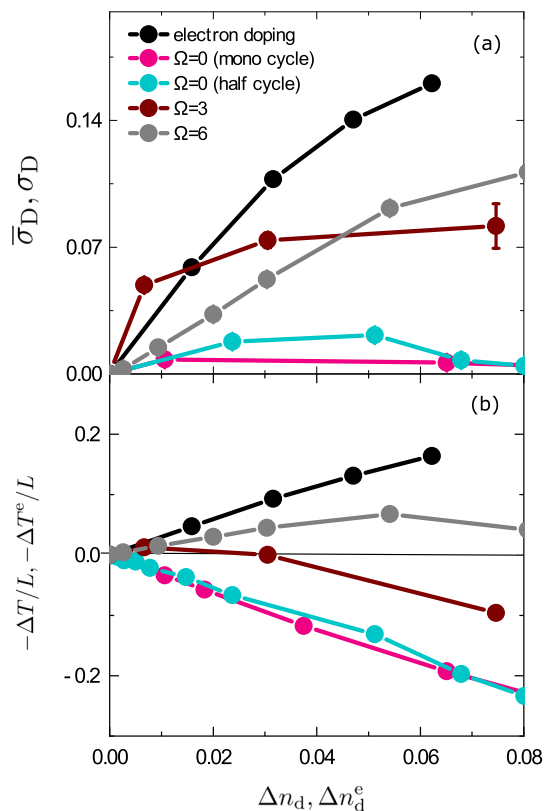


FIG. 3. (a) $\bar{\sigma}_D$ as the function of Δn_d and Δn_d^e . $\gamma = 0.4$ is taken. (b) $-\Delta T/L$ as the function of Δn_d and $-\Delta T^e/L$ as the function of Δn_d^e . All plots are obtained for the $L = 32$ IDEHM.

$\omega \neq 0$ due to a finite-size effect and its peak approaches $\omega = 0$ as L increases [70,89,90]. For $L = 32$, we can mask this finite-size effect by taking $\gamma = 0.4$. For half-cycle pulses, we obtain $\text{Re}[\sigma(\omega, \tau)]$ as shown in Figs. 2(e)–2(g). $|E(\omega)|$ given by $E(t) = E_0 e^{-(t-t_0)^2/(2t_d^2)} \cos[\Omega(t-t_0)]$ for $(\Omega, t_d) = (0, 4)$ is shown in Fig. 2(h). We obtain $\Delta n_d = 0.02, 0.07, \text{ and } 0.08$ for Figs. 2(e)–2(g), respectively. Even if we find finite σ_D as shown in Fig. 2(e) with small Δn_d , further increase in Δn_d does not enhance σ_D as shown in Figs. 2(f) and 2(g), but rather suppresses it.

The strong suppression of σ_D suggests that strong fields localize nonequilibrium states. When a thermal state with $\sigma_D \neq 0$ approaches an MBL state with $\sigma_D = 0$, σ_D is suppressed and the center of gravity of low-energy spectral weights shifts to higher energy [91–93], which is similar to the structure seen in Figs. 2(b), 2(c), 2(f), and 2(g) when E_0 is large. The suppression of the Drude weight is clearly shown in Fig. 3(a) if we compare $\bar{\sigma}_D$ (see below) induced by $\Omega = 0$ pulses (see magenta and light blue points) with those by photon absorption with $\Omega = 3$ (see brown points) and $\Omega = 6$ (see gray points) pulses as well as electron doping (see black points). Here, we introduce an time-averaged Drude weight $\bar{\sigma}_D = \frac{1}{2} \sum_{\tau=12,14} \int_{\omega=0}^{2\eta} d\omega \text{Re}\sigma(\omega, \tau)$ in Fig. 3(a) with $2\eta = 0.15$. Note that carrier density by electron doping are represented as $\Delta n_d^e = \frac{1}{2L} [\langle I \rangle_{\text{doped}} - \langle I \rangle_{\text{half}}]$, where $\langle \mathcal{O} \rangle_{\text{doped}}$ and $\langle \mathcal{O} \rangle_{\text{half}}$ are expectation values of \mathcal{O} for electron-doped and half-filled IDEHM, respectively. The factor $1/2$ is introduced to compare the carrier density of electron-doped

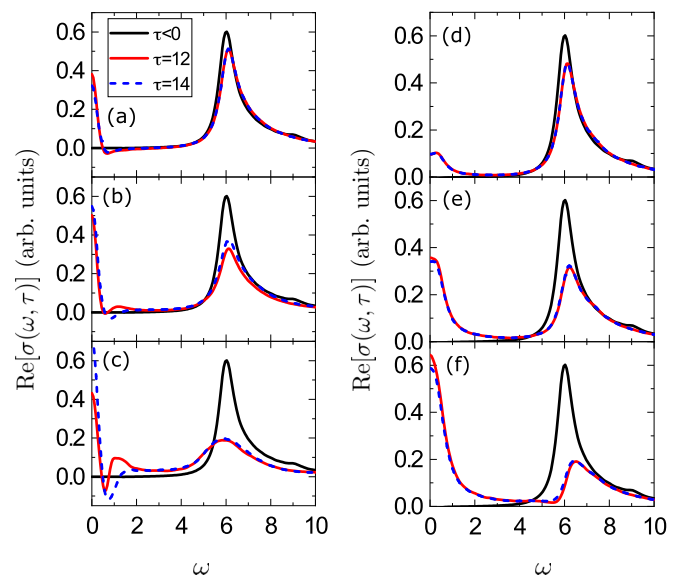


FIG. 4. $\text{Re}[\sigma(\omega, \tau)]$ excited by $\Omega = 3$ [$\Omega = 6$] pulses with (a) $E_0 = 0.9$, (b) $E_0 = 1.5$, and (c) $E_0 = 1.8$ [(d) $E_0 = 0.12$, (e) $E_0 = 0.24$, and (f) $E_0 = 0.36$]. Black, red, and blue-dashed lines are for $\tau < 0$, $\tau = 12$, and 14 , respectively. All plots are obtained by taking $\gamma = 0.4$ for the half-filled $L = 32$ IDEHM with $(U, V) = (10, 3)$.

systems with that of pulse-excited systems where the same number of holons and doublons are excited. We find that σ_D of electron-doped IDEHM (see Sec. S2 in the Supplemental Material [75]) has large values leading to $\sigma_D \propto \Delta n_d$. Upon electron doping, electrons are free to move and their kinetic energy decreases as indicated by black points in Fig. 3(b). The change of the kinetic energy for electron doped IDEHM is defined as $\Delta T^e = -t_h \sum_{j,\sigma} [\langle B_{j,\sigma} \rangle_{\text{doped}} - \langle B_{j,\sigma} \rangle_{\text{half}}]$. Upon electron doping, spectral weights above the Mott gap transfer to those at $\omega = 0$ due to spin-charge separation [94]. Since the change of total spectral weights is determined by $-\frac{1}{2L} \Delta T^e$ according to the optical sum rule [95], the decrease of kinetic energy contributes to the enhancement of σ_D . Photon absorptions also lead to metallic states following $\bar{\sigma}_D \propto \Delta n_d$. $\bar{\sigma}_D$ of IDEHM excited by $\Omega = 3$ and 6 pulses are obtained from $\text{Re}[\sigma(\omega, \tau)]$, which exhibits large spectral weights at $\omega = 0$ as shown in Fig. 4. $\Omega = 3$ pulses with $E_0 = 0.9$ [Fig. 4(a)], $E_0 = 1.5$ [Fig. 4(b)], and $E_0 = 1.8$ [Fig. 4(c)] lead to $\Delta n_d = 0.007, 0.03, \text{ and } 0.07$, respectively. $\Omega = 6$ pulses with $E_0 = 0.12$ [Fig. 4(d)], $E_0 = 0.24$ [Fig. 4(e)], and $E_0 = 0.36$ [Fig. 4(f)] lead to $\Delta n_d = 0.009, 0.03, \text{ and } 0.05$, respectively. Note that $\bar{\sigma}_D$ is affected by the emergence of spectral weights at $\omega \sim 0.5$ (see Sec. S3 in the Supplemental Material [75]).

In contrast to the electron-doped and photon-absorbed systems, there is no metallization when excitations are induced by a photon nonabsorbable $\Omega = 0$ pulse causing quantum tunneling. The change of kinetic energy $\Delta T = -t_h \sum_{j,\sigma} [\langle B_{j,\sigma} \rangle_t - \langle B_{j,\sigma} \rangle_0]$ induced by $\Omega = 0$ pulses exhibits a significant difference from other cases: $-\Delta T < 0$ monotonically decreases with increasing Δn_d as shown by magenta and light blue points in Fig. 3(b). We consider that a large increase in ΔT is associated with a restricted mobility due to the presence of strong fields, which leads to the strong suppression of $\bar{\sigma}_D$.

The time evolution of an entanglement entropy $S_E = -\sum_i p_i \ln p_i$ with the eigenvalue p_i of a reduced density matrix obtained by contracting half of the whole system shows different behavior when IDEHM is excited by quantum tunneling and by photon absorption (see Sec. S4 in the Supplemental Material [75]). For photon absorption, S_E shows rapid linear growth and saturates at the end of pulse irradiation. On the other hand, for quantum tunneling, S_E shows slow logarithmic growth and continues to grow slowly even after the end of pulse irradiation. The slow growth of S_E [96–101] is considered to be one of the manifestations of the localized nature of excited states by a high-field terahertz pulse.

Floquet effective Hamiltonians. We see how $\Omega = 0$ pulses localize nonequilibrium states in the 1D Mott insulator. For simplicity, we consider the dc limit of the Hamiltonian (1) with $V = 0$ taking $A(t) = \Delta t$. Using the Schrieffer-Wolff transformation [24,102], we obtain an effective model for resonant driving $U = p\Delta \gg t_h$, taking nonzero integers p . Due to the collective nature of the Zener breakdown, tunneling occurs not only between nearest-neighbor sites but also across several sites associated with $\Delta \leq U$ [34–39]. The $\xi \sim 1.5$ indicates that the dominant contribution to the breakdown is quantum tunneling within a few sites, which can be described as the effect of resonant electric fields with $\Delta = U/p$ for $p \lesssim 3$. The leading-order effective Hamiltonians for $p = 1, 2$, and 3 are

$$\begin{aligned}\mathcal{H}_{p=1}^{(0)} &= -t_h \sum_{j,\sigma} (h_{j+1,j,\sigma}^\dagger + h_{j+1,j,\sigma}), \\ \mathcal{H}_{p=2}^{(1)} &= \frac{t_h^2}{\Delta} [(T_1 + T_1^\dagger) - 2(T_2 + T_2^\dagger) + H_D^a - T_{XY}] \\ &\quad + \frac{t_h^2}{3\Delta} (H_D^b - T_3^b - T_{XY}), \\ \mathcal{H}_{p=3}^{(1)} &= \frac{t_h^2}{2\Delta} (H_D^a - T_{XY}) + \frac{t_h^2}{4\Delta} (H_D^b - T_3^b - T_{XY}),\end{aligned}$$

respectively (see Sec. S5 in the Supplemental Material [75]), where

$$\begin{aligned}h_{j,\sigma}^\dagger &= n_{j,-\sigma} (1 - n_{i,-\sigma}) c_{j,\sigma}^\dagger c_{i,\sigma}, \\ T_1 &= \sum_{j,\sigma} n_{j+2,-\sigma} (1 - n_{j,-\sigma}) (1 - 2n_{j+1,-\sigma}) c_{j+2,\sigma}^\dagger c_{j,\sigma}, \\ T_2 &= \sum_{j,\sigma} n_{j+2,\sigma} (1 - n_{j,-\sigma}) c_{j+2,-\sigma}^\dagger c_{j+1,-\sigma} c_{j+1,\sigma}^\dagger c_{j,\sigma}, \\ H_D^a &= \sum_{j,\sigma} n_{j+1,-\sigma} [-n_{j,\sigma} + 2n_{j+1,\sigma} (1 - n_{j,-\sigma})], \\ H_D^b &= \sum_{j,\sigma} n_{j,\sigma} [-n_{j+1,-\sigma} + 2n_{j,-\sigma} (1 - n_{j+1,-\sigma})], \\ T_3^b &= \sum_{j,\sigma} n_{j,\sigma} (1 - n_{j+2,-\sigma}) \\ &\quad \times (c_{j,-\sigma} c_{j+1,-\sigma}^\dagger c_{j+1,\sigma}^\dagger c_{j+2,\sigma} + \text{H.c.}), \\ T_{XY} &= \sum_{j,\sigma} [(1 - n_{j,-\sigma})(1 - n_{j,\sigma}) + n_{j+1,-\sigma} n_{j+1,\sigma}] \\ &\quad \times c_{j,-\sigma}^\dagger c_{j+1,-\sigma} c_{j+1,\sigma}^\dagger c_{j,\sigma}.\end{aligned}$$

The effective Hamiltonians suggest that the Floquet metastable states have conservations due to $[P + I, \mathcal{H}_{p=1}^{(0)}] = [P + 2I, \mathcal{H}_{p=2}^{(1)}] = [P, \mathcal{H}_{p=3}^{(1)}] = [I, \mathcal{H}_{p=3}^{(1)}] = 0$, where $P = \sum_k k n_k$ is the dipole moment. Since the resonance condition induces real excitations, the effect of a strong electric field remains in excited states even after a pulse disappears. Such conservation may break ergodicity and lead to exotic many-body dynamics. Indeed, it has numerically demonstrated that $\mathcal{H}_{p=1}^{(0)}$ can induce ergodicity-breaking many-body eigenstates [49] like quantum many-body scarring [103–115]. Also, dynamics governed by $\mathcal{H}_{p=2}^{(1)}$ is known to be nonergodic [44]. Kinetic constraints imposed by such conservation lead to the emergent fragmentation of the Hilbert space, generating exponentially many disconnected subspaces [44–53] even within a single symmetry sector. Dipole-moment-conserved system is a representative system with such restriction as seen in fractons [60–67], which localize charge excitations topologically. T_3^b included in $\mathcal{H}_{p=2}^{(1)}$ and $\mathcal{H}_{p=3}^{(1)}$ conserving both P and I is an example of showing doublon-assisted dipole-moment conserving processes, which does not produce Drude weight/superfluid density [116].

A strong $\Omega = 0$ pulse produce two effects in excited states: one is the injection of carriers promoting itinerancy, and the other is the restriction of motion promoting localization. As a result of their competing effects, the localization effect prevails in the $U \sim 10$ strong coupling region, and the excited states follow glassy dynamics [54–59] with weak-ergodicity breaking. We see the strong suppression of σ_D for $U = 7$ and 13 fixing $V/U = 0.3$ (see Sec. S6 in the Supplemental Material [75]). However, the suppression of σ_D for $U = 7$ is weaker than that for $U = 10$ and 13. This is because glassy states are unlikely to emerge in weak-coupling region, since the above discussion with the effective Hamiltonians is valid in strong-coupling regime. We note that the glassy state proposed in this Letter has a different origin from that induced by randomness near the Mott transition [117–122]. We expect that the glassy dynamics may be detected in ET-F₂TCNQ excited by a terahertz pulse with amplitude about 3.5 MV/cm.

Summary. We have investigated $\text{Re}[\sigma(\omega, \tau)]$ of pulse-excited states of the half-filled IDEHM using tDMRG. We have proposed that an insulator-to-glass transition is induced by strong mono- and half-cycle pulses, which leads to the suppression of σ_D . This is in contrast to the insulator-to-metal transition that occurs upon excitation by photon absorption accompanying $\sigma_D \propto \Delta n_d$. Restricted mobility due to strong fields induces glassy dynamics as seen in fracton systems. Not glassy but metallic states have been observed in the Mott insulator κ -(ET)₂Cu[N(CN)₂]Br excited by terahertz pulses in the experiment [11]. One possibility is that the enhancement of σ_D has been observed during electric field irradiation when nonequilibrium metastable states have not yet been reached (see Sec. S7 in the Supplemental Material [75]). Another possibility is that electron correlation is not so large that the subspaces in the fragmented Hilbert space are connected. Lastly, we note that qualitative differences in $\text{Re}[\sigma(\omega, \tau)]$ between photon absorptions and quantum tunnelings have recently been observed in a Mott insulator Ca₂RuO₄ [123].

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was carried out using computational resources of HOKUSAI at RIKEN Advanced Institute for Computational Science, the supercomputer system at the information initiative center, Hokkaido University, the facilities of the Supercomputer Center at Institute for Solid State Physics, the University of Tokyo, and supercomputer Fugaku provided by the RIKEN Center for Computational Science through the HPCI System Research Project (Project ID: hp170325, hp220048).

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