

Supplementary information for
Free electron topological bound state induced by light beam with
a twisted wavefront

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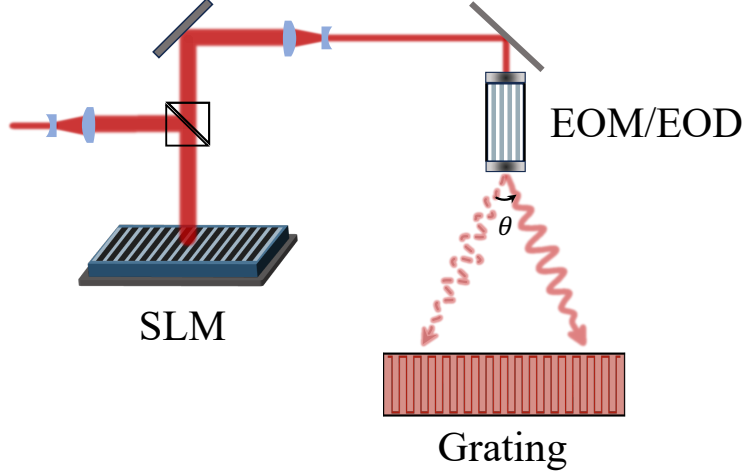
1. Experimental realization of twisted phase front using SLM

The realization of our proposed free-electron topological bound state relies on the synchronization between the slow travelling electron and the phase kink of the twisted light field. In the main text, we have presented a scheme using spatial light modulator (SLM) to modulate light field, as shown in Fig. 2. However, the response time of a typical SLM is on the order of microseconds, making it impractical to directly generate a phase kink that can keep up the travelling light.

In our setting (Supplementary Fig.1), therefore, we combine the SLM with a lens system and a high-speed electro-optic deflector (EOD) or electro-optic modulator (EOM), which is an additional illustration for Fig. 2 in the main text. The light beam is first modulated by the SLM and then reflected by a fixed mirror followed by an EOD/EOM driven by an ultrafast electrical signal, effectively decoupling the modulation and synchronization processes. Conceptually, the spatial phase pattern is pre-loaded onto the light beam through the SLM, and the EOD is then supposed to rapidly sweep this “kinked” beam across the grating to co-move with the electron. However, achieving this in practice poses formidable experimental challenges. As we acknowledge, there is a fundamental trade-off between the scanning speed and the spatial resolution (the number of resolvable points). While traditional acousto-optic deflectors (AODs) are fundamentally limited by the acoustic velocity and cannot phase-match with relativistic electrons, emerging EOD technologies—which have no acoustic inertia and offer deflection speeds limited only by electronics—provide a more promising theoretical pathway. Nevertheless, executing electro-optic signal conversion with the required femtosecond precision to perfectly overlap the hundred-femtosecond electron wavepacket with the optical phase kink remains highly challenging.

The angular velocity is required to precisely tune according to the far-field relation: $\omega_r = \beta c / l$, where βc is the group velocity of the electron pulse, and l is the distance from the EOD to the grating. In our setup, with typical values of $\beta = 0.02$ and a grating length of approximately $100 \mu m$, which is feasible for an angular velocity of $\omega_r = 10 \text{ MHz}$ and $l = 0.5 m$. Regarding the spatial extent of the phase kink and the beam diameter, maintaining a sufficient number of resolvable points while achieving ultra-high scanning speeds constitutes the primary technical bottleneck. A smaller optical beam diameter could theoretically increase the scanning speed, but it drastically reduces the spatial resolution, making it impossible to accurately encode the phase kink profile. Conversely, a larger beam diameter provides adequate resolution but

severely limits the sweep rate due to latency and temporal smearing. Therefore, while our proposed configuration serves as a theoretical blueprint, overcoming this essential trade-off will require future breakthroughs in high-bandwidth electro-optic technologies and advanced synchronization schemes.



Supplementary Fig.1: Conceptual setup for constructing a comoving twisted light wavefront using an electro-optic deflector (EOD) driven by an ultrafast signal. A light beam is split, with the reflected portion modulated by a spatial light modulator (SLM). The modulated beam is reflected back to the beam splitter, while the transmitted portion is conceptually guided onto an EOD. A lens system expands the modulated beam and a reduction lens system adjusts the phase kink size. The EOD is driven by an ultrafast RF/microwave source, aiming to reach synchronism between the light beam and electron pulses. Although this configuration theoretically imprints the twisted wavefront onto a grating and synchronizes it with the travelling electron, realizing the required scanning speed and spatial resolution simultaneously currently remains a significant experimental challenge.

2. TDSE simulation algorithm

In order to assess our tight-binding-approximated result of a two-level electron in the Bragg regime, we have to solve the time-dependent Schrödinger equation (TDSE) directly

$$i\hbar \frac{\partial}{\partial t} \psi(z, t) = \hat{H} \psi(z, t) = (\hat{H}_0 + \hat{H}_I) \psi(z, t), \quad (S1)$$

where the kinetic Hamiltonian of free electron is $\hat{H}_0 = E_0 + v_0(\hat{p} - p_0) + \frac{(\hat{p} - p_0)^2}{2\gamma^3 m_e}$. We choose the initial kinetic energy $\varepsilon_0 = 100 \text{ eV}$, corresponding to the electron velocity $v_0 = \beta c$ with the relative speed $\beta = 0.02$ and the Lorentz factor $\gamma = 1.001$. We take a realistic nanograting, whose longitudinal vector potential is $A(z, t) = -\frac{E_z}{\omega_L} \sin(\omega_L t - qz + \theta(z, t))$, with electric

field strength E_z , laser frequency ω_L , wavevector $q = \frac{2\pi}{\Lambda}$ for a grating with a period Λ , and the phase θ . Thus, the interaction Hamiltonian can be written as

$$\begin{aligned}\hat{H}_I &= -\frac{e}{2\gamma m_e} [\hat{p} \cdot A(z, t) + A(z, t) \cdot \hat{p}] \\ &= -\frac{e}{\gamma m_e} A(z, t) \cdot \hat{p} - i\hbar k_z A_0 \cos(\omega_L t - qz + \theta(z, t)) \\ &\simeq -\frac{eA_0}{\gamma m} \sin(\omega_L t - qz + \theta(z, t)) \cdot \hat{p},\end{aligned}\tag{S2}$$

where we disregard the second term under the approximation $p_0 \gg \hbar k_z$ (this will be demonstrated in the following), and $\theta(z, t) = -\frac{\pi}{2} \tanh(\frac{z-vt-z_0}{0.001})$.

We express the electron wavefunction as the slow-moving part with an initial phase:

$$\psi(z, t) = \chi(z, t) e^{-\frac{i(E_0 t - p_0 z)}{\hbar}}.\tag{S3}$$

Thus, the left-hand side (LHS) of Eq. (S1) is

$$i\hbar \frac{\partial}{\partial t} \psi(z, t) = e^{-\frac{i(E_0 t - p_0 z)}{\hbar}} \left(E_0 + i\hbar \frac{\partial}{\partial t} \right) \chi(z, t).\tag{S4}$$

Meanwhile, the right-hand side (RHS) becomes

$$\begin{aligned}& \left[e^{-\frac{i(E_0 t - p_0 z)}{\hbar}} \left(E_0 + v_0((\hat{p} + p_0) - p_0) + \frac{((\hat{p} + p_0) - p_0)^2}{2\gamma^3 m_e} \right) \right. \\ & \quad \left. - e^{-\frac{i(E_0 t - p_0 z)}{\hbar}} \frac{eA_0}{\gamma m_e} \sin(\omega_L t - qz + \phi_0) \cdot (\hat{p} + p_0) \right] \chi(z, t) \\ & \quad - \left[e^{-\frac{i(E_0 t - p_0 z)}{\hbar}} i\hbar k_z \frac{eA_0}{2\gamma m_e} \cos(\omega_L t - qz + \phi_0) \right] \chi(z, t) \\ & \simeq e^{-\frac{i(E_0 t - p_0 z)}{\hbar}} \left(E_0 + v_0 \hat{p} + \frac{\hat{p}^2}{2\gamma^3 m_e} - \frac{eA_0}{\gamma m_e} \sin(\omega_L t - qz + \theta(z, t)) \cdot (\hat{p} + p_0) \right) \chi(z, t),\end{aligned}\tag{S5}$$

here we omit the third term since $\frac{i\hbar k_z}{2} \cos(\omega_L t - qz + \phi_0) \ll p_0 \sin(\omega_L t - qz + \phi_0)$ under $p_0 \gg \hbar k_z$. Thus, we simplify the TDSE as

$$\begin{aligned}i\hbar \frac{\partial}{\partial t} \chi(z, t) &= \left(\frac{\hat{p}^2}{2\gamma^3 m} + \left[v_0 - \frac{eA_0}{\gamma m} \sin(\omega_L t - qz + \theta(z, t)) \right] \hat{p} \right) \chi(z, t) \\ &\quad - \frac{eA_0 p_0}{\gamma m} \sin(\omega_L t - qz + \theta(z, t)) \chi(z, t).\end{aligned}\tag{S6}$$

Substitute the momentum operator $\hat{p} = -i\hbar \frac{\partial}{\partial z}$ and $p_0 = \gamma\beta mc$, $A_0 = \frac{E_0}{\omega_L}$ yields

$$i\hbar \frac{\partial}{\partial t} \chi(z, t) = \left(-i\hbar \left[v_0 - \frac{eE_0}{\gamma m \omega_L} \sin(\omega_L t - qz + \theta(z, t)) \right] \frac{\partial}{\partial z} \right) \chi(z, t) - \left[\frac{\hbar^2}{2\gamma^3 m} \frac{\partial^2}{\partial z^2} + \frac{eE_0 \beta c}{\omega_L} \sin(\omega_L t - qz + \theta(z, t)) \right] \chi(z, t). \quad (S7)$$

The partial difference equation would be solved easier in the co-moving frame: $\zeta = z - v_0 t$, and $t' = t$, and we have

$$\begin{aligned} d\zeta &= dz - v_0 dt, & dz &= d\zeta + v_0 dt', \\ dt' &= dt, & dt &= dt', \end{aligned} \quad (S8)$$

which leads to the relations

$$\begin{aligned} \frac{\partial}{\partial z} &= \frac{\partial}{\partial \zeta} \frac{\partial \zeta}{\partial z} + \frac{\partial}{\partial t'} \frac{\partial t'}{\partial z} = \frac{\partial}{\partial \zeta}, \\ \frac{\partial}{\partial t} &= \frac{\partial}{\partial \zeta} \frac{\partial \zeta}{\partial t} + \frac{\partial}{\partial t'} \frac{\partial t'}{\partial t} = \frac{\partial}{\partial t'} - v_0 \frac{\partial}{\partial \zeta}. \end{aligned} \quad (S9)$$

In the co-moving frame, Eq. (S7) becomes

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \psi(z, t) &= i\hbar \left(\frac{\partial}{\partial t'} - v_0 \frac{\partial}{\partial \zeta} \right) \psi(\zeta, \tau_c) \\ &= \left(-i\hbar \left[v_0 + \frac{eE_0}{\gamma m \omega_L} \sin(q\zeta - \theta(\zeta)) \right] \frac{\partial}{\partial \zeta} \right) \chi(\zeta, \tau_c) \\ &\quad - \left[\frac{\hbar^2}{2\gamma^3 m} \frac{\partial^2}{\partial \zeta^2} - \frac{eE_0 \beta c}{\omega_L} \sin(q\zeta - \theta(\zeta)) \right] \chi(\zeta, \tau_c), \end{aligned} \quad (S10)$$

where $\theta(\zeta) = -\frac{\pi}{2} \tanh(\frac{\zeta - z_0}{0.001})$ and $\tau_c = ct'$. Divided by $\hbar c$ in both sides, we obtain

$$\begin{aligned} i \frac{\partial}{\partial \tau_c} \chi(\zeta, \tau_c) &= -i \left[\frac{eE_0}{\gamma mc \omega_L} \sin(q\zeta - \theta(\zeta)) \right] \frac{\partial}{\partial \zeta} \chi(\zeta, \tau_c) \\ &\quad - \left[\frac{\hbar}{2\gamma^3 mc} \frac{\partial^2}{\partial \zeta^2} - \frac{eE_0 \beta}{\hbar \omega_L} \sin(q\zeta - \theta(\zeta)) \right] \chi(\zeta, \tau_c). \end{aligned} \quad (S11)$$

Notice that all the variables are calculated in the length unit of μm and the time of fs . Assume that the electric field on the grating surface is $0.5 \times 10^7 V/m$ and the incident laser $\lambda_L = 0.2 \mu m$. Define

$$\alpha_1 = \frac{eE_0}{\gamma m_e c \omega_L} = 3.11 \times 10^{-7}, \quad \alpha_2 = \frac{\hbar}{2\gamma^3 m_e c} = 1.92 \times 10^{-7} \mu m, \quad (S12)$$

$$\alpha_0 = \frac{eE_0 \beta}{\hbar \omega_L} = 0.016 \mu m^{-1}.$$

The difference between the Bragg regime and Raman-Nath regime is determined by the factor

$$Q = \frac{\epsilon}{2|\kappa|} = \frac{\alpha_2 q^2}{\alpha_0} = \left(\frac{\hbar^2}{2m_e e c^3} \right) \frac{\omega_L^3}{\beta^3 \gamma^3 E_0}, \quad (S13)$$

For our concern, we choose the parameters $Q = 1.5$ to obtain a two-level electron Rabi oscillation. As a result, the simplified equation is

$$i \frac{\partial}{\partial \tau_c} \chi(\zeta, \tau_c) = -i\alpha_1 \sin(q\zeta - \theta(\zeta)) \frac{\partial}{\partial \zeta} \chi(\zeta, \tau_c) - \left[\alpha_2 \frac{\partial^2}{\partial \zeta^2} - \alpha_0 \sin(q\zeta - \theta(\zeta)) \right] \chi(\zeta, \tau_c). \quad (S14)$$

Then we apply the discretization for the spatial parameter ζ by dividing the spatial domain into N_ζ parts, with the interval $\delta\zeta = (\zeta_{max} - \zeta_{min})/N_\zeta$, which leads to the following,

$$\begin{aligned} \frac{\partial^2}{\partial \zeta^2} \chi(\zeta, t) &= \frac{1}{\delta\zeta^2} [\chi(\zeta + \delta\zeta, t) + \chi(\zeta - \delta\zeta, t) - 2\chi(\zeta, t)] \\ -i\alpha_1 \sin(q\zeta - \theta(\zeta)) \frac{\partial}{\partial \zeta} \chi(\zeta, t) &= \frac{\alpha_1}{\delta\zeta} \cos(q\zeta - \theta(\zeta)) \chi(\zeta, t) \\ &- \frac{\alpha_1}{2\delta\zeta} [e^{i(q\zeta - \theta(\zeta))} \chi(\zeta + \delta\zeta, t) + e^{-i(q\zeta - \theta(\zeta))} \chi(\zeta - \delta\zeta, t)]. \end{aligned} \quad (S15)$$

Thus, the discretized Hamiltonian becomes

$$\begin{aligned} i \frac{\partial}{\partial \tau_c} \chi(\zeta, \tau_c) &= - \left[\frac{\alpha_2}{\delta\zeta^2} + \frac{\alpha_1}{2\delta\zeta} e^{i(q\zeta - \theta(\zeta))} \right] \chi(\zeta + \delta\zeta, \tau_c) \\ &- \left[\frac{\alpha_2}{\delta\zeta^2} + \frac{\alpha_1}{2\delta\zeta} e^{-i(q\zeta - \theta(\zeta))} \right] \chi(\zeta - \delta\zeta, \tau_c) \\ &+ \left[\frac{2\alpha_2}{\delta\zeta^2} + \frac{\alpha_1}{\delta\zeta} \cos(q\zeta - \theta(\zeta)) + \alpha_0 \sin(q\zeta - \theta(\zeta)) \right] \chi(\zeta, \tau_c). \end{aligned} \quad (S16)$$

In order to compute the time evolution of the slow-varying part $\chi(\zeta, \tau_c)$ for a given initial state $\chi(\zeta, \tau_0)$, we define the vector wavefunction with N_ζ components

$$v(\tau) = \left(\chi(\zeta_1, \tau), \chi(\zeta_2, \tau), \dots, \chi(\zeta_{N_\zeta}, \tau) \right)^T. \quad (S17)$$

Then, the differential equation (S16) can be simplified as

$$i \frac{\partial}{\partial \tau} v(\tau) = H(\tau) v(\tau). \quad (S18)$$

Applying the time discretization for the interval $\tau - \tau_0$ by equally dividing it into N_t parts with the subinterval $\Delta\tau = (\tau - \tau_0)/N_t$ and we use an implicit Crank-Nicholson integrator to propagate the vector wavefunction from one time step to the next. The formal solution to Eq. (S18) can be expressed in terms of the time evolution operator

$$v(\tau + \Delta\tau) = U(\tau + \Delta\tau, \tau) v(\tau), \quad (S19)$$

where the time evolution operator can be expressed as

$$U(\tau + \Delta\tau, \tau) = (1 + i \Delta\tau H(\tau)/2)^{-1} (1 - i \Delta\tau H(\tau)/2). \quad (S20)$$

The solution of Eq. (S17) can give us the dynamics of the PINEM electron with an initial condition. The final wavefunction could be expressed as

$$v(\tau) = \prod_{n=1}^{N_t} U(\tau_0 + n\Delta\tau, \tau_0 + (n-1)\Delta\tau) v(\tau_0). \quad (S21)$$

3. The full phase-space profiles of the topological charge

In this study, we simulate an electron with an energy of approximately 100 eV and a group velocity of $v_0 = \beta c$, with $\beta = 0.02$. Synchronization between the phase velocity of the optical nearfield induced by a 200 nm laser is achieved using a nanograting with a period $\Lambda = 4$ nm. The initial electron state is presented as a superposition of two Gaussian wavepackets locating at $\hbar(k_0 + q/2)$ and $\hbar(k_0 - q/2)$ in the momentum domain, where $k_0 = \gamma m v_0 / \hbar$. This setting of the electron-light interaction lies in Bragg regime.

We simulate the interaction using the time-dependent Schrödinger equation (TDSE). The electron interacts with the light field for a duration of 25 ps, divided into discrete time steps. By recording the amplitude of the wave function at each time step, we obtain the temporal

profile of the topological charge's spatial distribution. A Fourier transform is performed to derive its momentum distribution.

Supplementary Fig.2a-d present simulation results, providing additional details for Fig. 3 in the main text. The initial electron state of 2a and 2b is prepared as a superposition of two sidebands following Jackiw-Rebbi solution (Eq. 9 in the main text):

$$\psi_{JR}(p) = \frac{1}{\sqrt{2\sqrt{2\pi}\sigma_p}} \left[\exp\left(-\frac{(p - \hbar q/2)^2}{4\sigma_p^2}\right) + i \exp\left(-\frac{(p + \hbar q/2)^2}{4\sigma_p^2}\right) \right], \quad (S22)$$

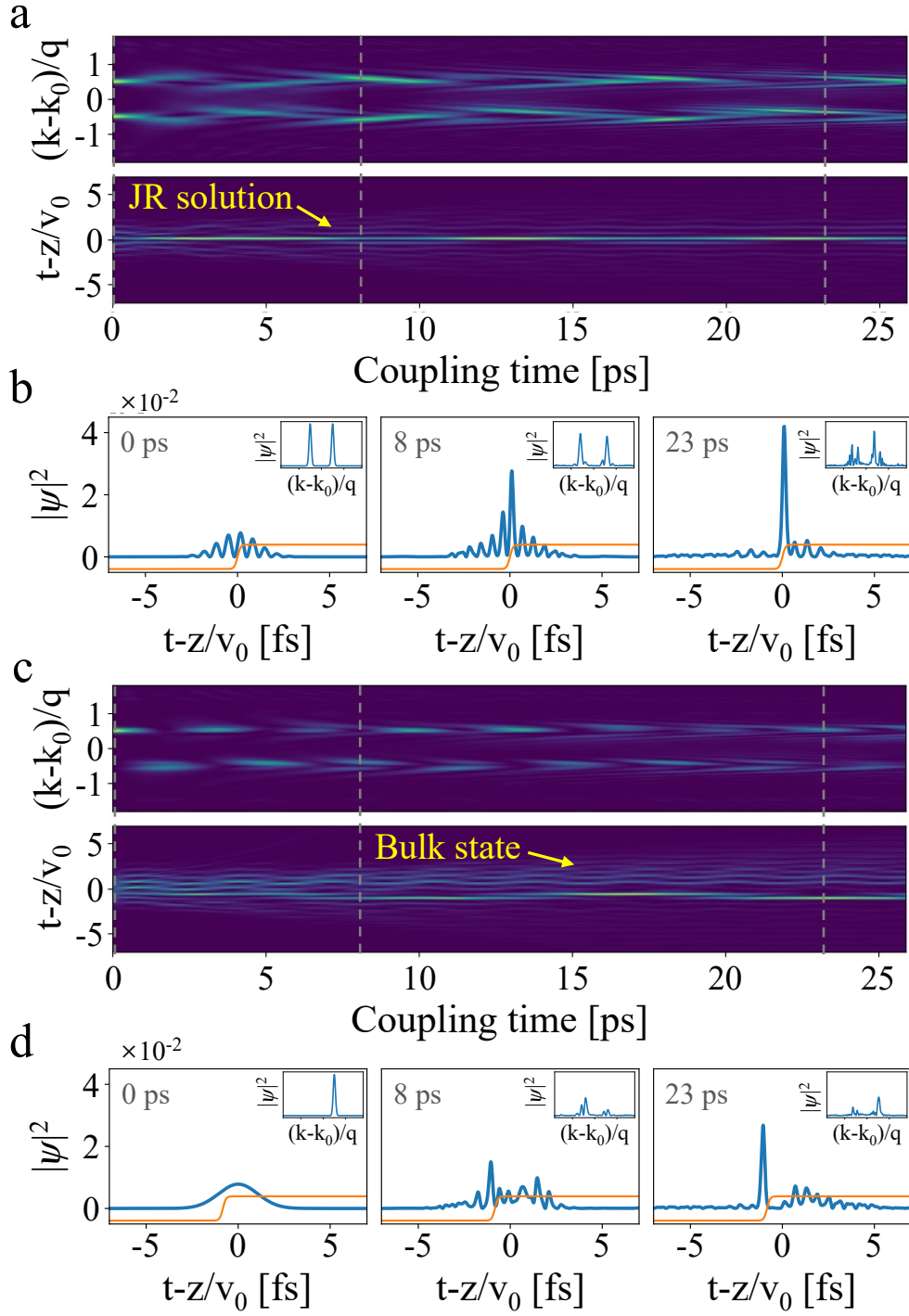
The topological bound state of Jackiw-Rebbi solution develops from this inital state, where the evolutions of both the temporal and momentum distributions are shown in Supplementary Fig.2a, and three time moments (0 *ps*, 8 *ps* and 23 *ps*) are presented in Supplementary Fig.2b for further clarity. The termportal distribution of the input eletron becomes localized around the phase kink (Supplementary Fig.2b), exhibiting a confinement feature that is immune to the intrinsic dispersion over time. By comparing the results in Supplementary Fig.2b and Supplementary Fig.2d, we see that the temporal profile of electron becomes almost fully trapped around the phase kink, indicating that the electron is primarily projected onto the topological bound state defined by Jackiw-Rebbi solution. Furthermore, the sidebands in momentum space (first row in Supplementary Fig.2a) split into small ones and flatten as time elapses, resulting in more complex structures that will not be expanded here. This provides additional details for Fig. 3a in the main text.

Supplementary Fig.2c and Supplementary Fig.2d, we present a simulation result of an initial electron prepared as a Gaussian wavepacket in both temporal and momentum domains, compared with Fig. 3b in the main text. The temporal envelop is offset by $\Delta t = 0.8fs$ from the phase kink, and the momentum envelop is set to a single sideband at $k_0 + q/2$, as shown in the first plot of Supplementary Fig.2d. The wavefunction can be written as

$$\psi_0(p) = \frac{1}{\sqrt{2\sqrt{2\pi}\sigma_p}} \exp\left(-\frac{(p + \hbar q/2)^2}{4\sigma_p^2}\right) \exp\left(i\frac{pv}{\hbar}\Delta t\right), \quad (S23)$$

The evolution of the electron wavepacket (Supplementary Fig.2d) shows the development of a localized bound state around the phase kink, similar to the evolution in Supplementary Fig.2b. However, a portion of the wavepacket gradually moves away from the kink. The non-trapped

part can be explained by the bulk states of the effective Dirac equation (Eq. 4 in the main text), which will be discussed later. This simulation demonstrate the robustness of the topological bound state induced by the optical phase kink. As long as there is overlap between the electron's temporal distribution and the phase kink, the electron is always projected onto the Jackiw-Rebbi solution. The evolution in momentum space (first row in Supplementary Fig.2c) exhibits characteristics of both Rabi oscillations, corresponding to the bulk state, and sideband splitting, corresponding to the topological bound state.



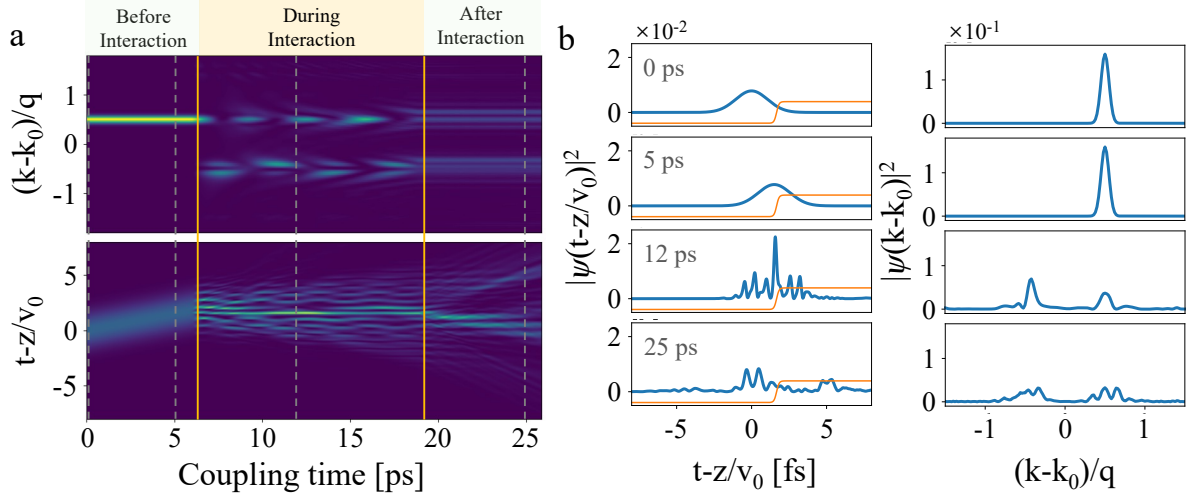
Supplementary Fig.2: TDSE simulation of the temporal profile of the topological bound state in both momentum and temporal domain. (a) the input electron state follows the form of Jackiw-Rebbi solution (Eq. 9 in the main text), exhibiting clear development of topological bound state. (c) the input electron state is a Gaussian wavepacket. (b)(d) the snapshots corresponding to the moments in (a)(c) marked by the grey dotted lines, emphasizing the development of the solitary state around the phase kink.

4. Simulation of a finite-regime interaction and robustness to detuning

We present a simulation of the interaction between a Gaussian electron wavepacket and a finite-region optical field with a twist wavefront. The simulation is divided into three parts due to the optical field, which is turned on at 6 fs and off at 19 fs, represented by the vertical orange line in Supplementary Fig.3a. This includes the time evolution of the wavepacket in both temporal and momentum domains. The corresponding distributions at four time points are shown in Supplementary Fig.3b. The initial electron is modeled as a Gaussian wavepacket, with its momentum envelop centered around $k_0 + q/2$ and its temporal envelop located at the center of comoving frame (shown in the first row of Supplementary Fig.3b).

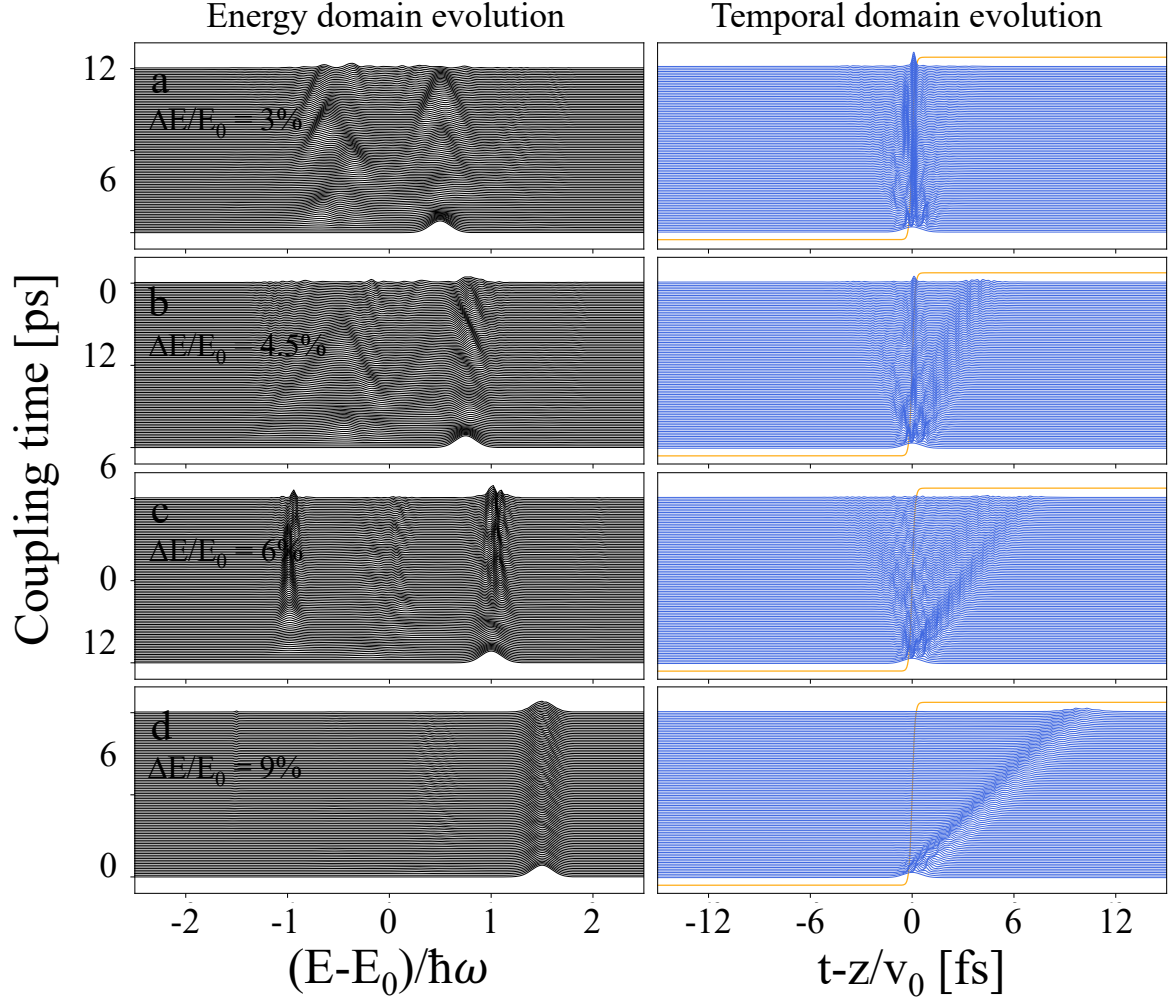
As shown in Supplementary Fig.3a, the electron propagates freely in the first regime, where the momentum distribution remains unchanged and the temporal distribution shifts slightly in the comoving temporal domain due to small detuning, as seen in the second row of Supplementary Fig.3b. When the optical field turns on at 6 fs, we position the twisted phase front to spatially overlap with the temporal profile of the electron. This spatial overlap guarantees a non-zero projection of the electron's state onto the topological bound state, resulting in a localized spatialtemporal distribution, as shown in the third row of Supplementary Fig.3b. The momentum distribution mainly occupies $k_0 + q/2$ and $k_0 - q/2$. Similar to the result in Supplementary Fig.2c, 2d, there are some extended waves representing bulk states away from the kink. At around 19 fs, the optical field is turned off, and the electron propagate freely again. In the absence of the twisted optical field, the solitary property vanishes, and the temporal distribution splits (fourth row in Supplementary Fig.3b) due to its momentum distribution corresponding to two distinct group velocities.

For clarification, we made a video (see the attached file) based on the simulation of Supplementary Fig.3, showing the evolution of both temporal and momentum profile of a Gaussian-input electron in a finite-regime optical field.



Supplementary Fig.3: simulation of QEW evolution in a finite-region optical field. The input electron state is a Gaussian wavefunction in both momentum and spatial domain, where the center of momentum wavepacket locates at $k_0 + q/2$, shown in the first line of (b). The first regime is the free-evolution in vacuum, where the electron exhibits slippery in the comoving frame. The optical near-field on grating is marked by the orange line, enable the electron-light interaction. The electron pulse enters the region at $t = 6 \text{ ps}$ and leaves at $t = 19 \text{ ps}$, the interaction with the near-field forms a topological state of Jackiw-Rebbi solution due to spatial overlap with the twist phase front, exhibiting a localized spatial distribution shown in (b). When electron leaves the grating region, the topological state exhibits strong dispersion in free-evolution, and splits into two parts in spatial domain.

In order to specify that how much detuning between the electron energy and the phase-front velocity is tolerable to maintain the validity of the Jackiw-Rebb solution, we include simulation results in Supplementary Fig.4 for detuning from 3% to 9%. The detuning is defined as ratio between shifted energy $\Delta E = E - E_0$ and the resonant energy E_0 with optical near-field ($E_0 = 100 \text{ eV}$ in our setup, e.g., $\Delta E = 3 \text{ eV}$ for 3% detuning). The detuning is implemented by offsetting the initial energy distribution of the QEW from E_0 in energy domain in our simulation. These plots show that the bound state persists for detuning from 1.5% to 4.5%, with gradually reduced coupling strength out of this region. This quantifies the tolerance and demonstrates the robustness of our scheme.



Supplementary Fig. 4: TDSE Simulation of Detuned Quantum Electron–Light Interaction. We present TDSE simulations exploring how detuning modifies the interaction between a QEWS and an optical near-field. The time evolution of the electron’s energy envelope (left column) and temporal envelope (right column) are shown for energy detunings of (a) 3 eV, (b) 4.5 eV, (c) 6 eV, and (d) 9 eV. Detuning is implemented by offsetting the initial energy distribution from the resonance energy E_0 . Formation of a Jackiw–Rebbi (JR) solution requires the electron sidebands to lie at $\pm\hbar\omega/2$, corresponding to $\pm 3\%$ energy detuning. (a) At 3 eV detuning, the QEWS exhibits pronounced temporal localization at the optical phase kink, indicative of a well-formed JR bound state. (b) For 4.5 eV detuning ($\sim 4.5\%$), the energy distribution deviates slightly from $\hbar\omega/2$, barely preserving a synthetic two-level structure. The temporal profile retains spatial-temporal localization but shows dispersion, implying coexistence of JR features and wavepacket broadening. (c) At 6 eV detuning ($\sim 6\%$), the initial energy sideband locates at $\hbar\omega$, and light–electron coupling generates additional sidebands at 0 and $-\hbar\omega$. The resulting temporal envelope undergoes significant spreading, indicating the collapse of the JR solution. (d) With 9 eV detuning, the electron–light coupling is negligible. The energy distribution, initially centered at $1.5\hbar\omega$, remains unchanged, and the temporal envelope exhibits a unidirectional drift in the co-moving frame, indicating misalignment between the electron’s temporal structure and the optical phase kink.

5. Bulk modes and zero-energy mode

In this section, we analyze and simulate the bulk and zero-energy modes base on the constructed Dirac equation in Eq. 4 of the main text. Initially, we ignore the quadratic term δk^2 assuming $\delta k < q$. The bulk modes can be derived by solving the normal Dirac Hamiltonian without the kink-induced mass term:

$$H_D(\delta k) = -\frac{\hbar^2 q}{2m} \delta k \cdot \sigma_z - |\kappa| \sigma_x = -\eta \delta k \cdot \sigma_z - |\kappa| e^{i\theta} \sigma_x. \quad (S24)$$

Here, the phase profile $\theta = 0, \pi$, is a constant. The eigenvalues of this Hamiltonian are

$$E_{\pm}(\delta k) = \pm \sqrt{|\kappa|^2 + \eta^2 \delta k^2}. \quad (S25)$$

The corresponding eigenstates for bulk states can be written as

$$v_+(\delta k) = \frac{1}{\sqrt{2|E_+(\delta k)|}} \begin{pmatrix} -e^{i\theta} \sqrt{E_+(\delta k) - \eta \delta k} \\ |\kappa| \\ \sqrt{E_+(\delta k) - \eta \delta k} \end{pmatrix}, \quad v_-(\delta k) = \frac{1}{\sqrt{2|E_-(\delta k)|}} \begin{pmatrix} e^{i\theta} \sqrt{\eta \delta k - E_-(\delta k)} \\ |\kappa| \\ \sqrt{\eta \delta k - E_-(\delta k)} \end{pmatrix}. \quad (S26)$$

In our model, the spinor is composed of the sidebands of a two-level electron, and δk is a continuous variable defined by the electron's momentum spread. It represents the pseudo-momentum space, independent of the synthetic lattice composed of sidebands. The dominant contribution comes from $\delta k = 0$, and we can obtain the eigenvectors for bulk modes as shown in the table below.

	$\theta = 0$	$\theta = \pi$
E_+	$v_+ = \frac{1}{\sqrt{2}} \begin{pmatrix} -1 \\ 1 \end{pmatrix} \equiv \downarrow\rangle$	$v_+ = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \equiv \uparrow\rangle$
E_-	$v_- = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \equiv \uparrow\rangle$	$v_+ = \frac{1}{\sqrt{2}} \begin{pmatrix} -1 \\ 1 \end{pmatrix} \equiv \downarrow\rangle$

Supplementary Table 1. the eigenvector derived from effective Dirac Hamiltonian for $\theta = 0, \pi$.

The simulation of the bulk state is shown in Supplementary Fig.5 for the electron temporal wavefunction, which is located within the optical field with a constant phase $\theta = \pi$. The initial momentum wavefunction for the bulk state is given as a superposition of the two sidebands according to the eigenvector of the effective Dirac equation:

$$\begin{aligned}
\psi^+(p) &= \frac{1}{\sqrt{2\sqrt{2\pi}\sigma_p}} \left[\exp\left(-\frac{(p - \hbar q/2)^2}{4\sigma_p^2}\right) + \exp\left(-\frac{(p + \hbar q/2)^2}{4\sigma_p^2}\right) \right] e^{i\frac{pv_0}{\hbar}\Delta t}, \\
\psi^-(p) &= \frac{1}{\sqrt{2\sqrt{2\pi}\sigma_p}} \left[\exp\left(-\frac{(p - \hbar q/2)^2}{4\sigma_p^2}\right) - \exp\left(-\frac{(p + \hbar q/2)^2}{4\sigma_p^2}\right) \right] e^{i\frac{pv_0}{\hbar}\Delta t},
\end{aligned} \tag{S27}$$

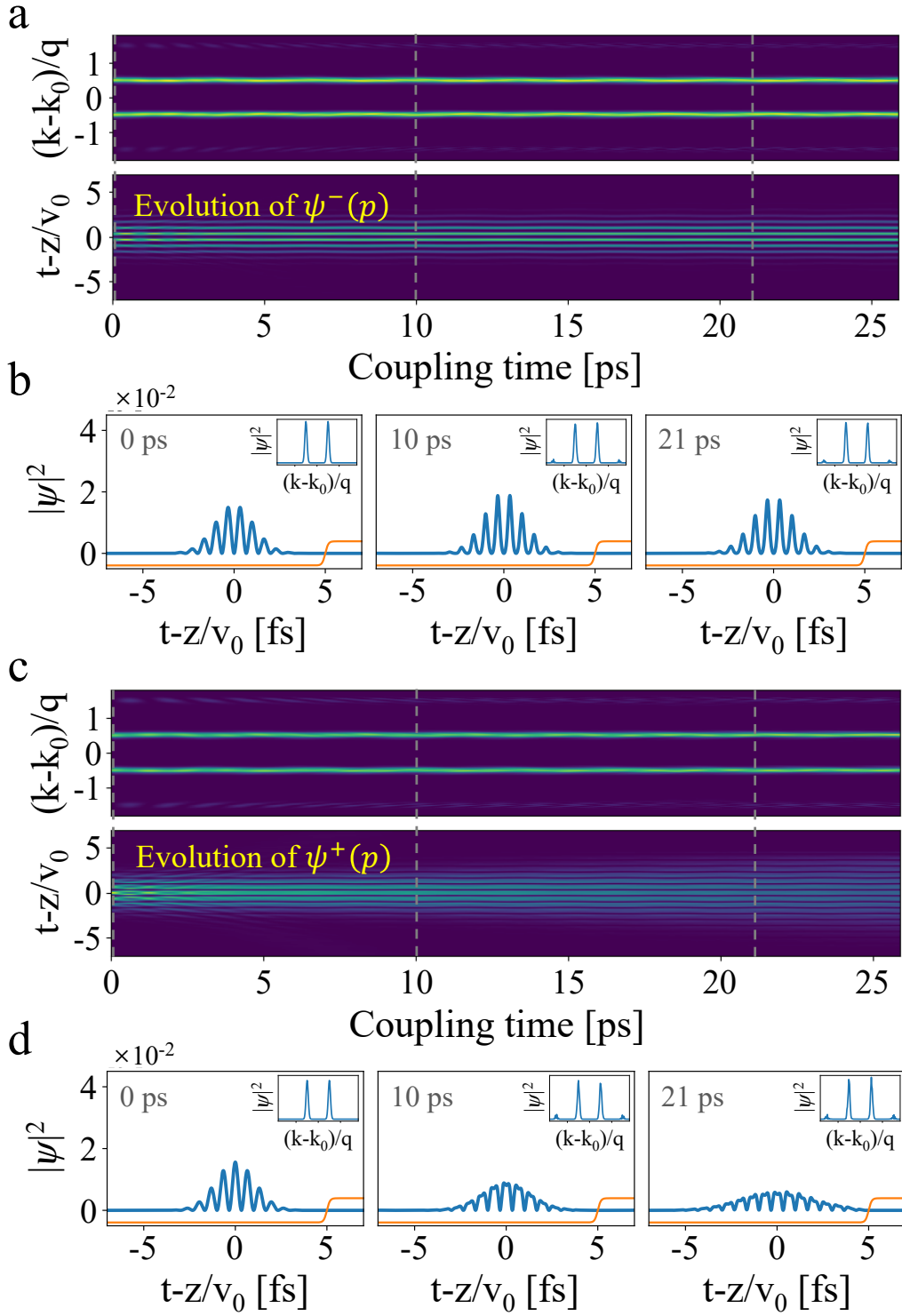
where $\psi^+(p)$ and $\psi^-(p)$ correspond to the positive and negative eigenvalues, respectively. The additional phase term $e^{i\frac{pv_0}{\hbar}\Delta t}$ ensures that the temporal center of the electron is offset (about 5 fs) from the kink, as shown in Supplementary Fig.5c and Supplementary Fig.5d. The time evolution of the negative energy bulk state is presented in the left column, while the positive energy bulk state is shown on the right. For both eigenenergies, the electron's momentum distribution remains unchanged during evolution (the first row of Supplementary Fig.5a and Supplementary Fig.5b), indicating that they are eigenstates of the system. However, the spatiotemporal distributions of the two eigenstates differs. The temporal envelop of positive-energy state becomes much wider than that of the negative-energy state, indicating a much stronger chirping.

The difference in chirping of the propagation between the negative-energy and positive-energy states can be explained by considering the neglected δk^2 term in Eq. 4. In our case, the pseudo-momentum term is much smaller than the coupling term $\eta\delta k \ll |\kappa|$, so the eigenenergy in Eq. S25 can be expanded to second order in δk : $E_{\pm}(\delta k) = \pm\sqrt{|\kappa|^2 + \eta^2\delta k^2} \simeq \pm|\kappa| \left(1 + \frac{\eta^2}{2\kappa^2}\delta k^2\right)$, the total energy is:

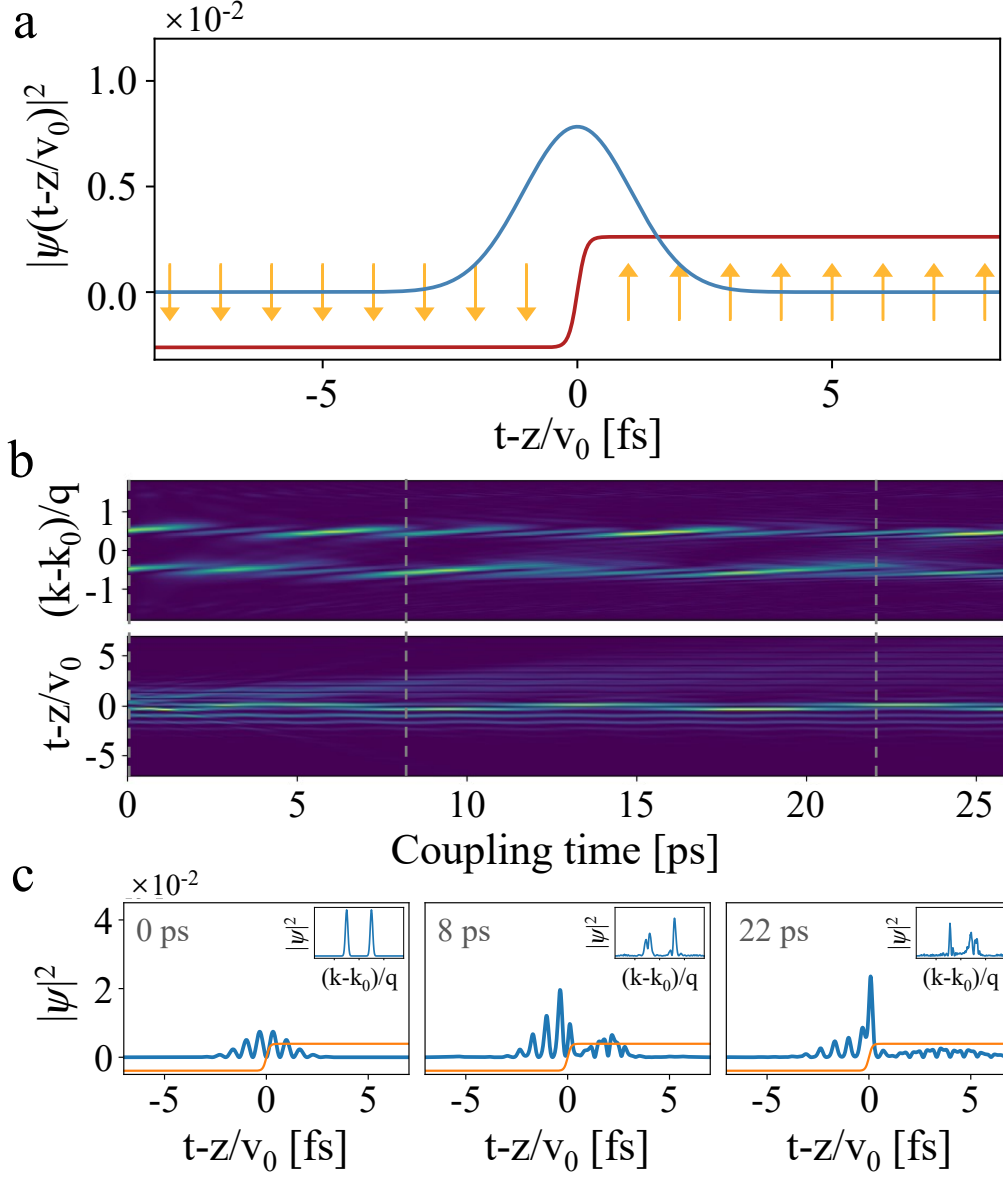
$$E_{\pm}^{tot} = \frac{\hbar^2\delta k^2}{2m} \pm |\kappa| \left(1 + \frac{\eta^2}{2\kappa^2}\delta k^2\right) = \frac{\hbar^2\delta k^2}{2m} \left(1 \pm \frac{\hbar^2 q^2}{4m|\kappa|}\right) \pm |\kappa|, \tag{S28}$$

where the parameter $\frac{\hbar^2 q^2}{4m|\kappa|} = 1.46$ in our simulation. The pseudo-momentum δk originates from the kinetic energy of the free electron, leading to the chirping. Therefore, the positive-energy bulk state exhibits a much stronger chirping behavior in the spatial domain than the negative-energy bulk state.

An interesting point, as shown in Table 1, is that the eigenvector of the negative energy for $\theta = 0$ and $\theta = \pi$ are orthogonal to each other. The same is true for the positive-energy state. If we define the up-spin state $|\uparrow\rangle \equiv \frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ 1 \end{pmatrix}$ and down-spin state $|\downarrow\rangle \equiv \frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ -1 \end{pmatrix}$, an electron wavepacket spatially overlapping with a phase kink is illustrated in Supplementary Fig.6a. The phase kink behaves like a domain wall, which defines distinct eigenstates on either side. Therefore, as long as the temporal envelop of electron overlaps with the kink, there will be non-zero projection of the electron state onto the topological bound state induced by the domain wall, resulting in a localized spatiotemporal distribution around the twist phase front. This is demonstrated in Fig. S6b, where the input electron is a bulk state with negative eigenenergy while its temporal envelop center coincides with the phase kink.



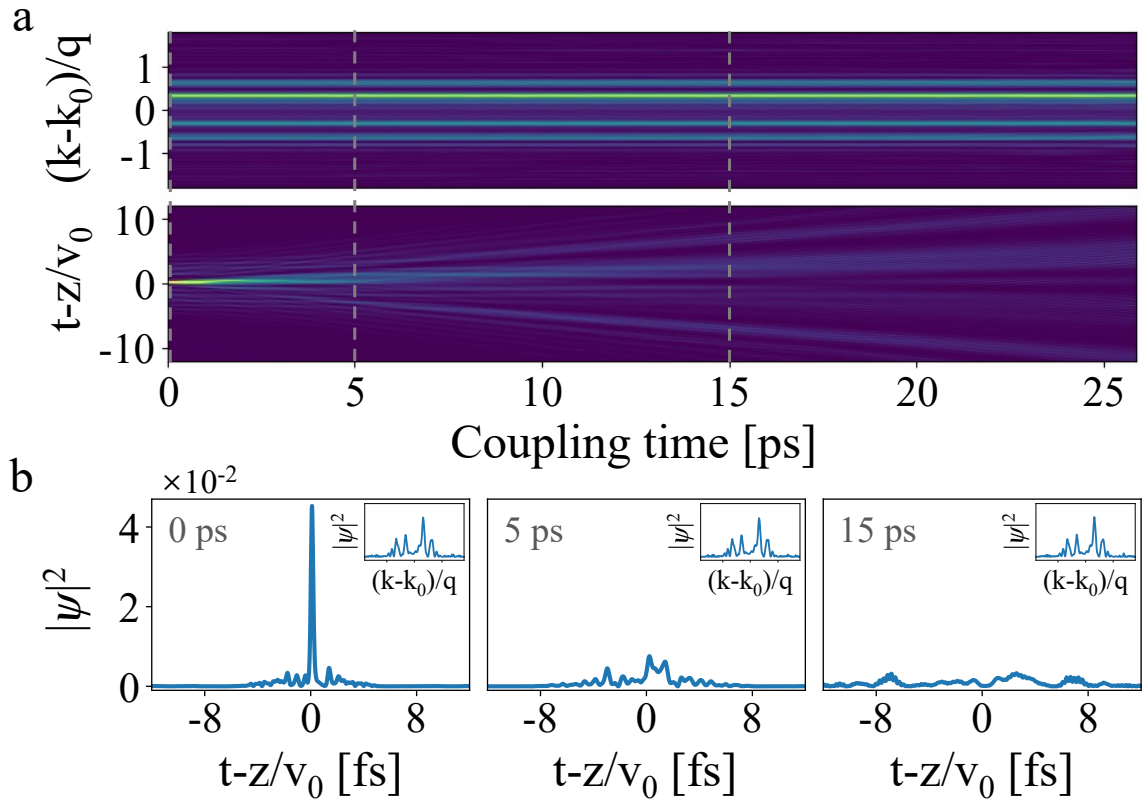
Supplementary Fig.5: Evolution of bulk state defined by the effective Dirac equation. The input electron state is set to be the eigenstates corresponding to positive and negative energies given in Eq. S28. The evolution of QEW's momentum and spatial wavefunction is shown in (a) (b), and the state at three time points marked by dashed grey line in evolution is shown in (c)(d).



Supplementary Fig.6: (a) Illustration of twist phase front domain wall for electron-light interaction. (b) time evolution of input electron state ψ^- in Eq. S27 with temporal overlap between the electron and phase kink, exhibits projection of bulk state onto the topological bound state, demonstrating the robustness of Jackiw-Rebbi solution. (c) The temporal and momentum distribution of three time points marked by dashed grey lines in (b), demonstrating formation of bound state and bulk modes.

6. Evolution of a JR electron in free space after turning off the twisted light beam

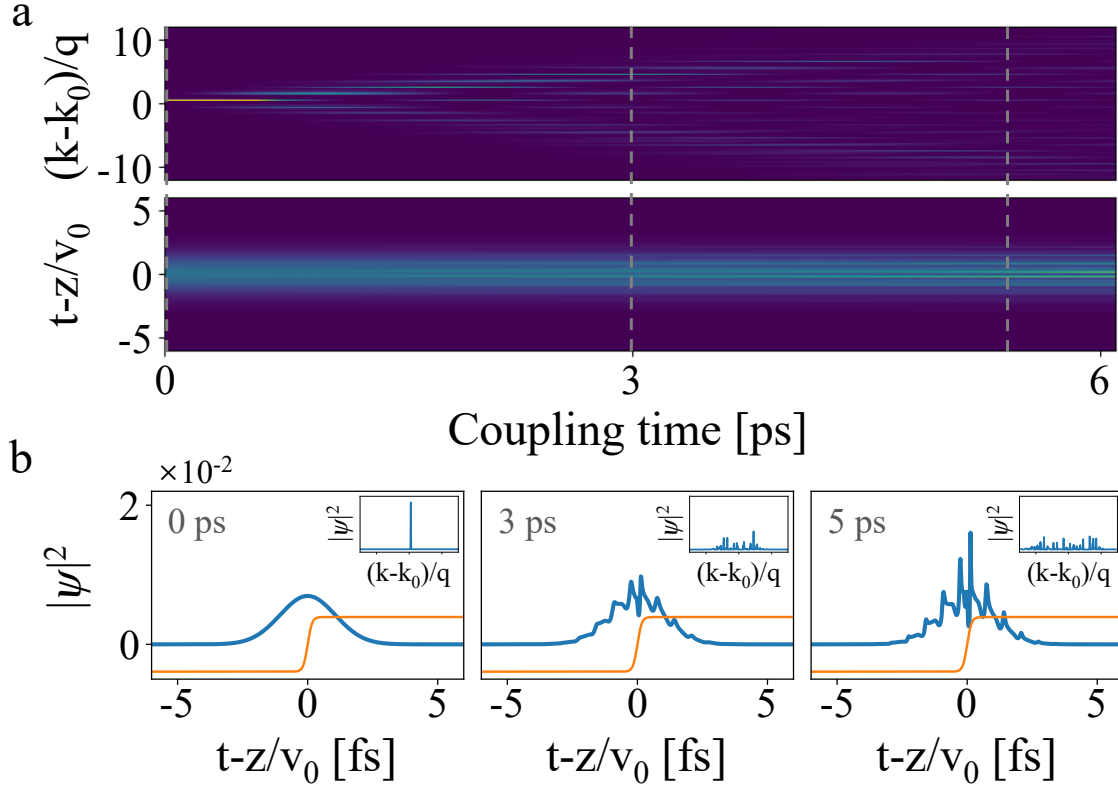
The topological properties of the free-electron state emerge from its hybridization with the twisted light field, meaning its stability depends on the presence of the light field. To explore this further, we present a TDSE simulation of the free evolution of a free-electron topological bound state after the light field is turned off, as shown in Supplementary Fig.7. The input electron used in the simulation is numerically derived from the result in Fig. 3a of the main text. The simulation reveals that the free-electron topological bound state transitions into a standard PINEM electron state and loses its topological nature. During the evolution, the spatiotemporal distribution undergoes both rapid spreading due to strong dispersion and splitting caused by the distinct velocities of the two PINEM sidebands.



Supplementary Fig.7: TDSE simulations of the evolution of the free electron topological bound state in vacuum when the twisted light beam is turned off. The time evolution of the temporal and momentum wavefunction is shown in (a), with the momentum distribution remaining unchanged and the temporal distribution experiencing significant dispersion over time. The input electron is prepared as a topological bound state derived from the numerical calculation in Fig. 3a in the main text, shown in the first plot in (b) with a localized temporal distribution and two split PINEM sidebands in the momentum domain. The three figures in (b) show the temporal distribution flattening and splitting into two parts due to dispersion, corresponding to the three time points marked by the grey-dotted lines in (a).

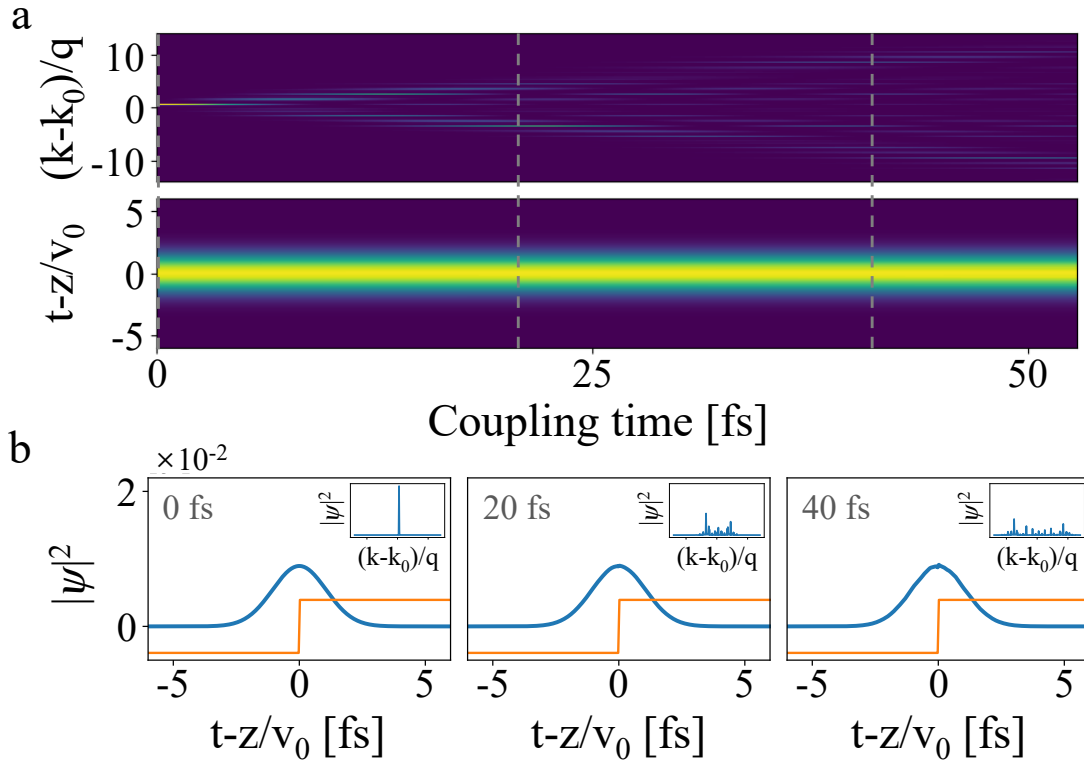
7. TDSE simulation without kinetic term in Bragg regime

In our theory, the effective Dirac equation (Eq. 4 of the main text) is derived from the Schrodinger equation describing the electron-light interaction in the Bragg regime. The spinor structure, consisting of two PINEM sidebands, arises due to the strong dispersion relative to the coupling strength between the electron and light field. Moreover, the dispersion term introduces a pseudo-momentum term $\delta k \sigma_z$, which accounts for the finite spread of the electron wavepacket and behaves similarly to a momentum-dependent chemical potential. To verify this, we present an additional TDSE simulation of the electron-light interaction in the absence of kinetic terms (α_2 in Eq. S14), as shown in Supplementary Fig.8. The simulation demonstrates that the formation of the spinor structure and the topological bound state disappears, while normal PINEM behavior occurs. Therefore, the kinetic term is incorporated into all results and conclusions in our work.



Supplementary Fig.8: TDSE simulation of a slow electron interacting with an optical field without the kinetic term. (a) Time evolution of the momentum and temporal wavefunction. (b) Temporal distributions at 0, 3 and 5 ps, with the corresponding momentum distributions shown in the insets. The simulation is conducted under the same condition as Fig. 3b in the main text, except for the omission of the kinetic term (α_2 in Eq. S14).

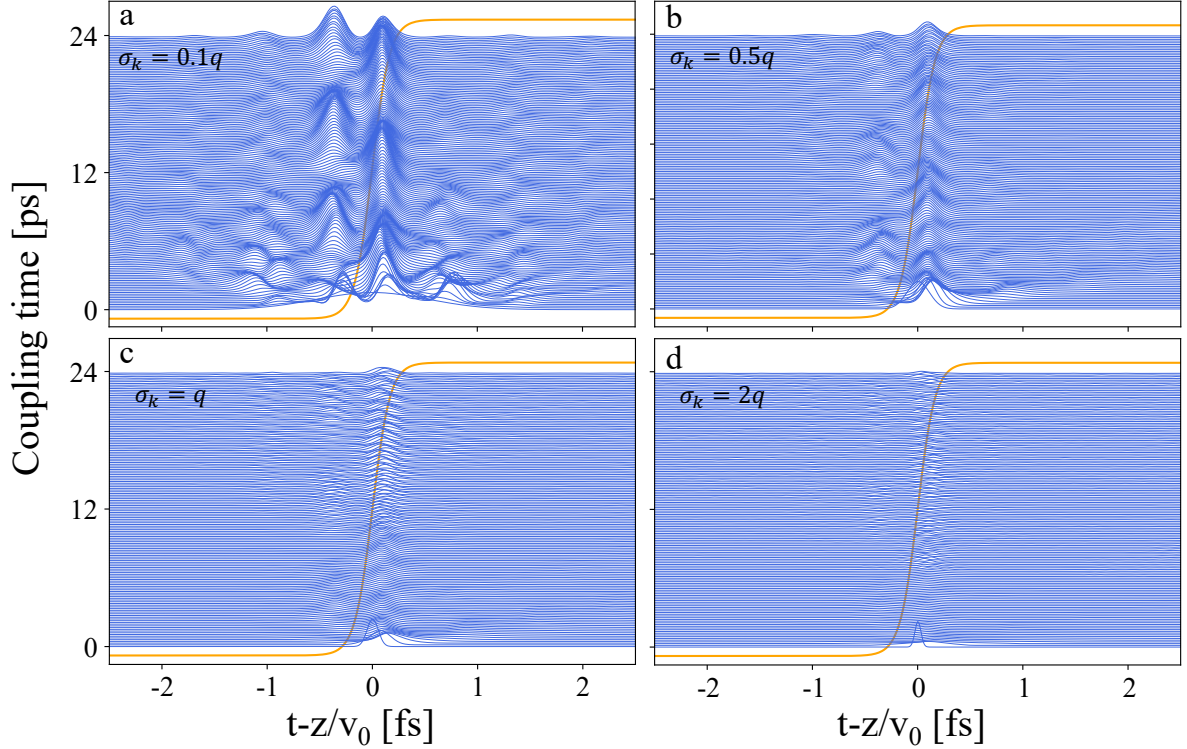
8. Interaction between a swift electron and a twisted light field



Supplementary Fig.9: Interaction between a swift electron and a twisted light field. (a) Time evolution of the momentum and temporal wavefunction for a swift electron (with energy set to 200 keV) interacting with the twisted light field. (b) Temporal distributions at 0, 20 and 40 fs, with the corresponding momentum distributions shown in the insets. The simulation exhibits normal PINEM behavior with increasing sidebands in momentum space, consistent with the Raman-Nath regime, and no free-electron topological bound state is formed under these conditions.

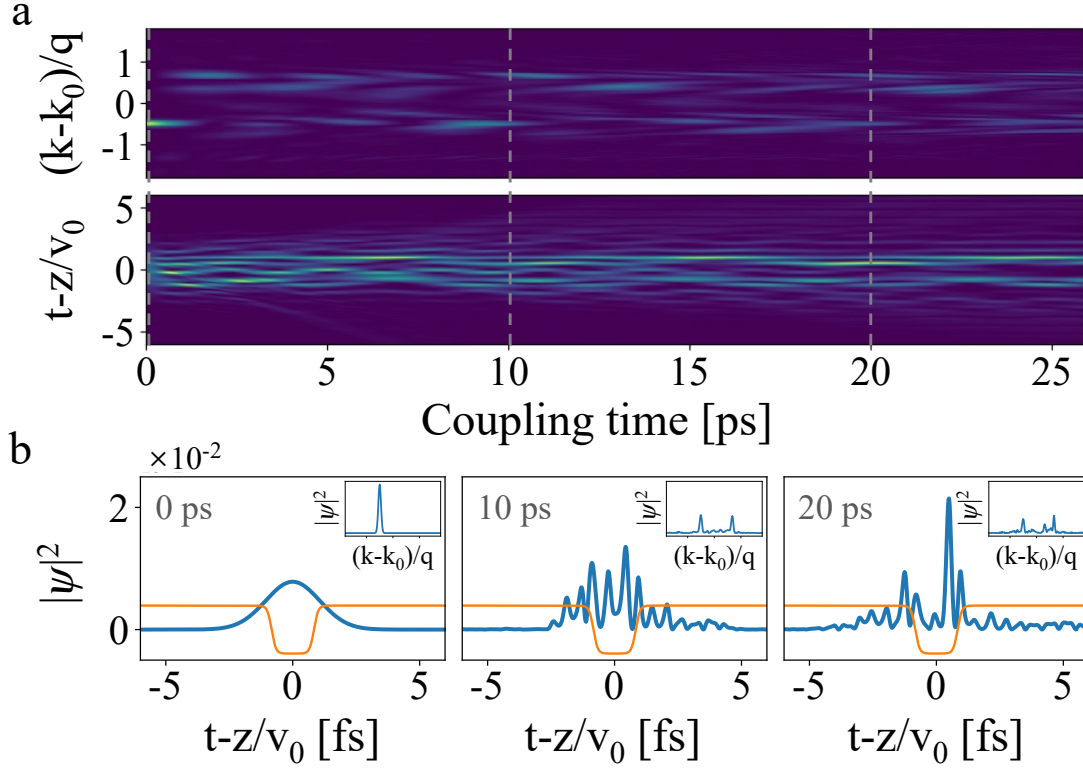
9. QEW-size dependent interaction with kink-loaded optical field

To clarify the dependence of the Jackiw-Rebbi solution for a free electron on the wavepacket size, we present TDSE simulations of four cases— including three typical regimes: $\sigma_k \ll q$, $\sigma_k \sim q$ and $\sigma_k \gg q$, where σ_k is the momentum spread of QEW while q is the photon wave vector—as shown in Supplementary Fig.10. These results show that the non-dispersive bound state still survives when $\sigma_k \approx q$, albeit with a slightly broader linewidth, while a point-like electron ($\sigma_k \gg q$, Supplementary Fig.10d) exhibits no evidence of spatial localization. These results confirm that our phenomenon is robust under the condition $\sigma_k < q$.



Supplementary Fig.10: TDSE simulation of QEW interactions with a kink-loaded optical field for varying QEW sizes. The width of optical phase kink is fixed at 1nm (0.17fs). From (a) to (d), the QEW shortens in temporal duration. As the temporal size becomes shorter, the spatial-temporal localization degrades. In (a) and (b), the electron's temporal duration is larger than optical cycle, hence the momentum sidebands remain well-seperable, allowing JR solution valid and the temporal localization persists. In (c), the temporal duration is comparable to both the optical cycly (also close to the kink width), causing the momentum sidebands overlap. Therefore, the modelling of JR solution breaks down in this regime and the temporal localization is weak. In (d), the QEW's temporal duration is much smaller than both the phase kink width and optical period, and no localization is observed.

10. Splitting electron wavepacket by a kink-antikink pair of light wavefront



Supplementary Fig.11: Generation of a pair of topological charges via a light beam with a kink-antikink phase front. (a) Time evolution of the momentum and temporal wavefunction for a Gaussian wavepacket overlapping with both kink and antikink phase fronts. (b) Temporal distributions at 0, 10 and 20 ps, with the corresponding momentum distributions shown in the insets. The simulation results show that the input electron becomes partly localized by the kink and antikink phase profiles, providing evidence of pair generation at the quantum-mechanical level.