

Electron-induced nonlinear dynamics in atomic chains

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We study the nonlinear response of collective optical resonances in linear atomic chains with metallic, semiconducting, and topologically insulating character to low-energy free electrons. The nonlinearity, which manifests in the amplitude and frequency of resonant features in cathodoluminescence and electron energy-loss spectra, is shown to depend on the speed and trajectory of the excitation as well as the length and electronic structure of the chain. Time-domain analysis of charge carrier dynamics within the atomic chain reveals that the Fermi velocity sets the threshold speed for triggering an electron-induced nonlinear response, a phenomenon which can elucidate nonlinear light-matter interactions on the nanoscale.

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Energetic electrons offer precise spatial and spectral characterization of subwavelength optical excitations supported in a plethora of nanophotonic platforms [1]. In typical electron spectroscopy experiments, focused electrons moving at relativistic speeds deposit energy in a sample that can be measured directly in electron energy-loss spectroscopy (EELS) [2–7], or in cathodoluminescence (CL) light emission [8–11], to map optical resonances on nanometer and meV scales [12]. Synchronization of electron beams with impinging optical pulses, such as in photon-induced near-field electron microscopy [13], enables exploration of dynamical processes governing nanoscale light-matter interactions [14–16]. Quantum mechanical properties encoded in free electrons and revealed in the light generated by their interaction with matter are actively explored in the context of quantum light sources and coherent control [17,18].

Light-matter interactions stimulated by moving electrons are characterized by the temporal and spatial overlap of the transient evanescent fields emanating from the electron beam with the target structure, such that the coupling strength depends on the electron trajectory and speed. Low-energy electrons thus afford longer light-matter interaction times that are anticipated to probe the nonlinear dynamics of material excitations [12,19]. In particular, ~ 100 eV electrons can deliver optical fluences comparable to those of the intense ultrashort optical pulses routinely employed in nonlinear optical experiments, and are predicted to produce spectral shifts and saturation of resonant features in CL and EELS associated with plasmons—the collective oscillations of conduction

electrons—supported by doped graphene nanoislands [20], while electron-induced nonlinear phenomena are anticipated to emerge in the excitation of few-level bosonic and fermionic systems [21,22]. The electron-induced nonlinear response of collective optical resonances in condensed matter systems has yet to be observed in experiment, for which the ideal electron excitation characteristics and target material properties remain to be established.

The Su-Schrieffer-Heeger (SSH) model—introduced to describe charge transport in polyacetylene chains [23]—constitutes a simple but versatile platform to explore topological and band structure effects in a wide range of physical phenomena. In the context of light-matter interactions, the SSH model offers insight used to characterize the collective nature of optical resonances in condensed matter systems [24–26], or to elucidate the role of electronic band structure in high-harmonic generation by intense ultrashort optical pulses [27,28]. The ability of the SSH model to capture synergies between electronic structure and optical excitations in finite chains constitutes an important resource in nanoscale nonlinear optics [29], where subwavelength optical resonances intensify the intrinsic nonlinear response of a material to electromagnetic fields.

Inspired by the paradigm of nonlinear optical response to low-energy electrons proposed in Ref. [20] for plasmons in graphene nanoislands, we theoretically explore electron-induced nonlinearities in finite one-dimensional atomic chains described by a SSH tight-binding model. Time-domain simulations of a swift electron interacting with collective electronic excitations in metallic, semiconducting, and topologically insulating (TI) SSH chains reveal a nonlinear response that manifests in the amplitude and frequency of resonant features in CL emission and electron energy-loss spectra. The electron-induced nonlinear response is found to exhibit an involved dependence on the intrinsic electronic properties of the atomic chain as well as the speed and trajectory of the impinging electron. Analysis of the dynamical induced charge density

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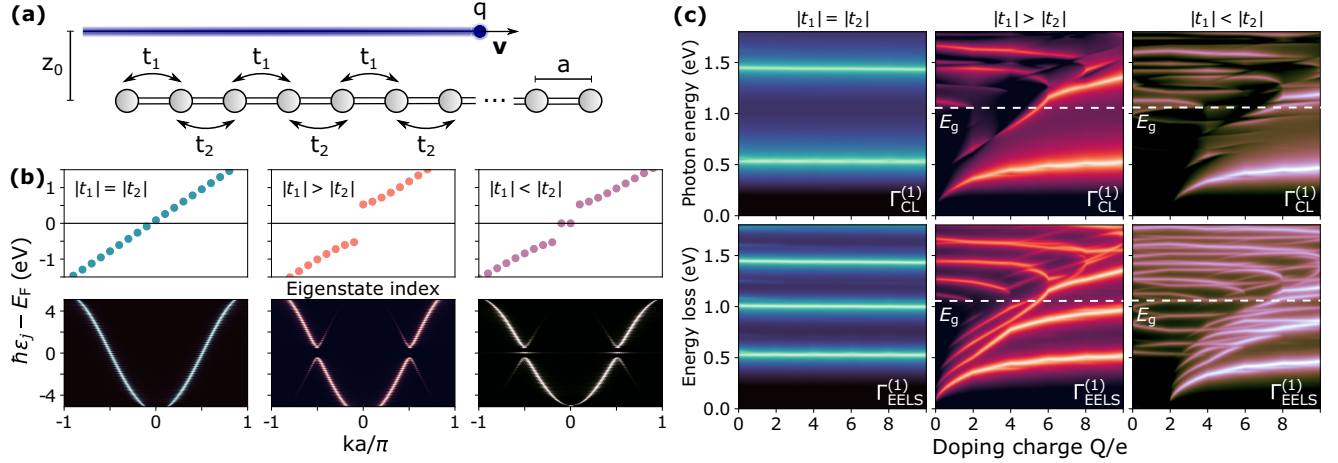


FIG. 1. Electron excitation of linear atomic chains. (a) Illustration of a SSH chain, composed of atoms arranged along $\hat{x}$ with uniform spacing a and described by a tight-binding Hamiltonian with alternating hopping energies t_1 and t_2 , excited by a particle with charge q moving at speed v parallel to the chain a distance z_0 above it. (b) Eigenenergy spectrum (top panels) and the Fourier transformed eigenstates $|a_{jk}|^2 L_j(\hbar\epsilon)$ weighted by Lorentzian functions L_j of width $\hbar\gamma$ centered on $\hbar\epsilon_j$ (bottom panels) for metallic (left column), semiconducting (middle column), and TI (right column) chains with $N = 100$ sites. (c) CL (upper row) and EELS (lower row) probabilities (on a logarithmic scale) predicted in linear response theory for an electron moving at speed $0.01c$ a distance $z_0 = 0.5$ nm above $N = 100$ chains doped with charge Q . Dashed lines indicate the band gap energy E_g .

and redistribution of population among single-electron energy eigenstates within SSH chains establishes the Fermi velocity of charge carriers as the threshold speed at which an external electron triggers a nonlinear response in plasmon resonances. Our findings elucidate the origin of electron-induced nonlinear light-matter interactions, which constitute an underexplored phenomenon that can probe nonlinear optical phenomena on atomic length scales.

The electronic properties of a finite one-dimensional atomic chain are described by the SSH tight-binding Hamiltonian

$$\mathcal{H}^{\text{TB}} = t_1 \sum_{l=1}^{N/2} |2l\rangle \langle 2l-1| + t_2 \sum_{l=1}^{N/2-1} |2l+1\rangle \langle 2l| + \text{H.c.}, \quad (1)$$

where $|l\rangle$ are single-electron orbitals located at the N atomic sites with coordinates x_l and t_i are alternating nearest-neighbor hopping energies [see Fig. 1(a)]. A discrete spectrum of electronic energies $\hbar\epsilon_j$ are obtained from the eigenvalue equation $\mathcal{H}^{\text{TB}} |j\rangle = \hbar\epsilon_j |j\rangle$, where states $|j\rangle = \sum_l a_{jl} |l\rangle$ are expanded in atomic site orbitals weighted by real-valued coefficients a_{jl} . The two hopping energies determine the electronic properties of the chain: As shown in Fig. 1(b), equal hoppings $|t_1| = |t_2|$ yield a metallic state, $|t_1| > |t_2|$ produces a semiconducting state, and $|t_1| < |t_2|$ results in a TI state with two edge states inside the gap. In this Letter, we fix $a = 0.1421$ nm and $t_2 = -2.8$ eV, which are the respective carbon-carbon spacing and nearest-neighbor hopping energy between carbon p orbitals in graphene [30], while choosing $t_1 = t_2 \pm 0.5$ eV to probe the semiconducting (+) and topologically insulating (−) states. The corresponding energy-momentum spectra of the SSH chains, obtained by decomposing the eigenstates in plane waves with wave vector k according to $a_{jk} = \sum_l a_{jl} e^{-ikx_l}$, reveal variations in electron velocity near

the band edges of “gapped” (i.e., semiconducting and TI) chains (see Supplemental Material [31]).

Adopting the formalism of previous studies [20,29], electron dynamics within the atomic chain are governed by a single-particle density matrix ρ that satisfies the equation of motion

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [\mathcal{H}^{\text{TB}} - e\phi, \rho] - \frac{\gamma}{2} (\rho - \rho^{(0)}), \quad (2)$$

where ϕ is the total electrostatic potential acting on the chain and γ is a phenomenological damping rate at which the density matrix relaxes to its equilibrium value $\rho^{(0)}$. In the site basis, the scalar potential $\phi_l = \phi_l^{\text{ind}} + \phi_l^{\text{ext}}$ is composed of an induced potential $\phi_l^{\text{ind}} = \sum_{l'} v_{ll'} \rho_{l'}^{\text{ind}}$ that describes self-interaction of the induced charge density ρ_l^{ind} mediated by the Coulomb interaction $v_{ll'}$ between electrons at sites l and l' , for which we use parameters associated with carbon $2p$ orbitals [32], and an external potential $\phi_l^{\text{ext}} = q|x_l \hat{x} - \mathbf{r}_0 - \mathbf{v}t|^{-1}$ that describes a point particle with charge q and velocity $\mathbf{v}$ passing through $\mathbf{r}_0 = (x_0, y_0, z_0)$ at time $t = 0$. The nonlinearity originates in Eq. (2) from the quadratic dependence on ρ introduced by the potential ϕ_l through the induced charge density $\rho_l^{\text{ind}} \equiv -2e(\rho_{ll} - \rho_{ll}^{(0)})$, where the factor of 2 accounts for spin degeneracy and the equilibrium state $\rho_{ll'}^{(0)} = \sum_j f_j a_{jl} a_{j'l'}$ is constructed from occupation factors f_j of states $|j\rangle$. The scalar potential ϕ treats induced and external interactions within the quasistatic approximation, while the form of ϕ^{ext} implies nonrelativistic particle speeds that are assumed constant, i.e., neglecting recoil effects due to the interaction of the charged particle with the atomic chain [20]. To focus on trends in the nonlinear response, we ignore possible screening of both the external and induced potentials by the dielectric environment.

Direct integration of Eq. (2) yields the charge density $\rho_l^{\text{ind}}(t)$ produced by a particle impinging on the atomic chain from an arbitrarily large distance; the spectral response is

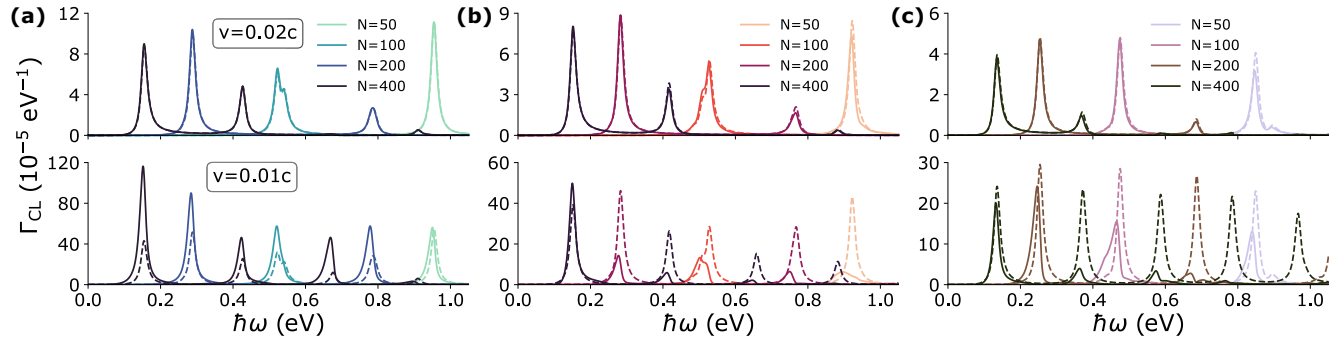


FIG. 2. Nonlinearity in the CL emission from atomic chains. CL spectra produced by electrons moving at speeds of $v = 0.02c$ (upper panels) and $v = 0.01c$ (lower panels) parallel to and 0.5 nm away from (a) metallic, (b) semiconducting, and (c) topologically insulating (TI) SSH chains composed of N sites and doped with $Q/e = N/10$ ($Q/e = N/10 + 2$ for the TI) electrons. Solid and dashed curves indicate the full and linear response, respectively.

analyzed from the Fourier components $\rho_{\omega,l}^{\text{ind}} = \int dt \rho_l^{\text{ind}}(t) e^{i\omega t}$. We compute the photon emission probability (photons emitted per unit time and frequency range) as $\Gamma_{\text{CL}} = 2\omega^3 |\mathbf{p}_\omega|^2 / 3\pi \hbar c^3$, where $\mathbf{p}_\omega = \sum_l x_l \rho_{\omega,l}^{\text{ind}}$ is the induced dipole moment; the EELS probability for an electron moving in a trajectory parallel to the chain is obtained as [20]

$$\Gamma_{\text{EELS}}^{\parallel} = -\frac{2q}{\pi \hbar v} \sum_l K_0 \left(\frac{\omega}{v} \sqrt{y_0^2 + z_0^2} \right) \text{Im} \{ \rho_{\omega,l}^{\text{ind}} e^{-i\omega x_l/v} \}, \quad (3)$$

where K_0 is a modified Bessel function.

To assess the nonlinear response, we compare the response obtained from time-domain simulations with that predicted in perturbation theory. Linearizing Eq. (2) in the frequency domain, we compute the induced charge in the random-phase approximation from $\rho_\omega^{\text{ind}} = \chi^{(0)}(1 - \chi^{(0)}v)^{-1} \phi_\omega^{\text{ext}}$ for matrices indexed by l , where

$$\chi_{ll'}^{(0)} = \frac{2e^2}{\hbar} \sum_{jj'} (f_{j'} - f_j) \frac{a_{jl} a_{j'l'} a_{j'l} a_{j'j}}{\omega + i\gamma/2 - (\epsilon_j - \epsilon_{j'})} \quad (4)$$

is the noninteracting susceptibility and the Fourier component of the external potential is $\phi_{\omega,l}^{\text{ext},\parallel} = (2q/v) K_0 [(\omega/v) \sqrt{y_0^2 + z_0^2}] e^{i\omega x_l/v}$ for a parallel trajectory (i.e., $\mathbf{v} = v\hat{\mathbf{x}}$) [20].

The linear response of SSH chains composed of $N = 100$ sites is characterized in Fig. 1(c), where CL and energy-loss probabilities are presented in the upper and lower panels, respectively, for electronic phases indicated by the hopping energy t_1 . In particular, CL and EELS probabilities are calculated for an electron passing with speed $0.01c$ a distance 0.5 nm away and parallel to the SSH chain as its doping charge Q is varied continuously, i.e., the occupation factors f_j defining the equilibrium state are populated by successively filling the lowest-energy unoccupied state until a total of $2N + Q/e$ electrons are distributed (assigning equal populations to degenerate states). In all cases, the response is dominated by prominent low-energy spectral features corresponding to plasmon resonances, with EELS presenting additional features associated with dark modes that do not radiate but absorb energy from the passing electron. The plasmon resonances of the metallic chain in Fig. 1(c) (left column) are less affected by doping, while plasmon modes in the semiconductor

(middle column) and TI (right column) emerge below the electronic band gap (E_g) for finite Q and gradually blueshift as the doping increases, contrasting features appearing above E_g due to individual electron-hole pair excitations (dressed by the Coulomb interaction) that become quenched due to Pauli blocking. The plasmonic features of the metallic chain are rendered relatively doping independent by the equally spaced electronic levels and abundance of free electrons that can undergo collective intraband transitions, while such resonances only become available in the gapped chains when states above the band gap are populated, becoming more prominent in the CL and EELS signals as more charge carriers are introduced. In contrast to the semiconducting chain, additional plasmon features appearing in the spectra of the TI chain are attributed to edge states within the band gap, which introduce additional excitation channels and offset the doping at which the conduction band is populated. Note that the resonances in gapped chains converge towards those in the metallic chain with increased doping.

Nonrelativistic electrons moving near an atomic chain undergo prolonged interactions with its supported optical resonances that manifest as a nonlinear response, which we study in Fig. 2. We compare perturbative (dashed curves) and time-domain (solid curves) simulations of CL emission from SSH chains of varying length interacting with a particle of charge $q = -e$ moving at speeds of $0.02c$ and $0.01c$. To provide a balanced comparison among metallic, semiconducting, and TI chains, we consider the same doping density $Q/e = N/10$ in the metallic and semiconducting chains while $Q/e = N/10 + 2$ in the TI (two charges populate edge states). The large but consistent doping density leads to well-defined plasmon resonances of similar frequency in all chains.

The qualitative behavior of the nonlinear response and the trend with chain length differ strikingly in the three electronic phases: In the metallic chain, CL emission from plasmons in Fig. 2(a) is enhanced compared to the linear spectrum and smoothly increases with N , while the plasmon resonances of gapped semiconducting and TI chains exhibit saturation. Edge states in the TI chains play a quantitative role, leading to an overall reduced CL signal compared to that of the semiconducting chains. The degree of saturation is similar between the two gapped systems, although the lower-order

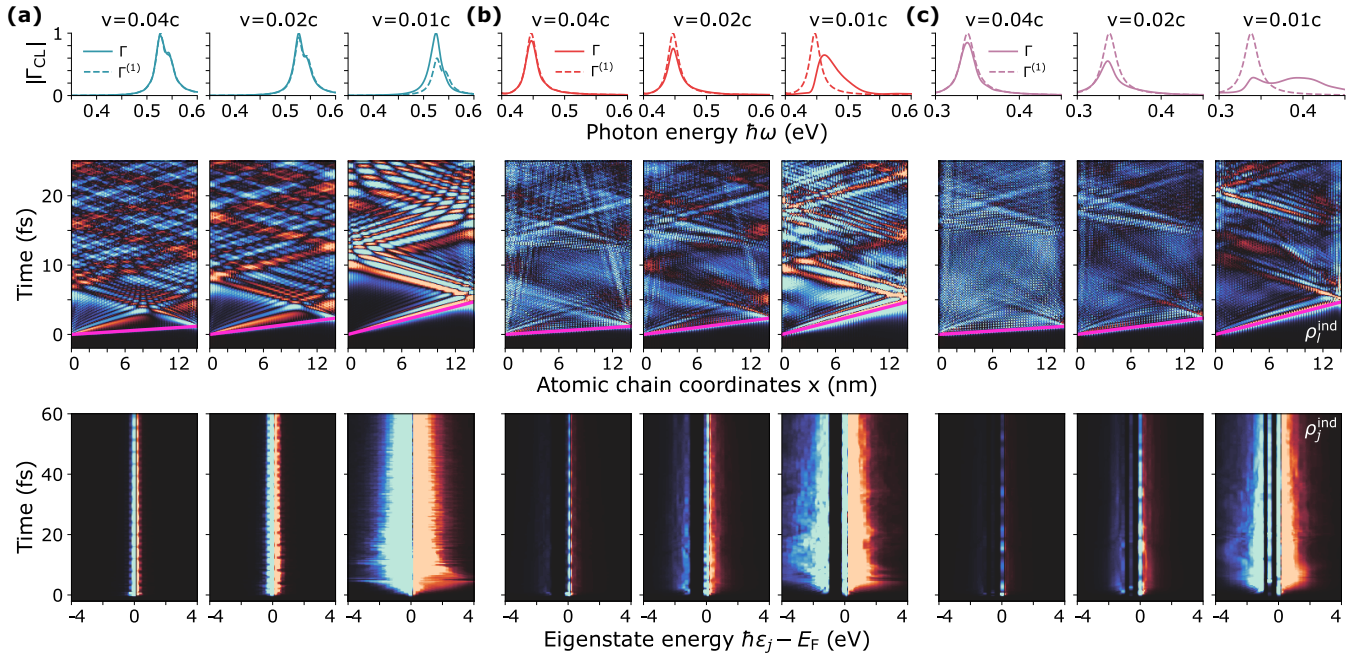


FIG. 3. Real-space charge carrier motion and population dynamics in atomic chains. We consider SSH chains with $N = 100$ sites and (a) metallic, (b) semiconducting, and (c) topologically insulating character that are doped with $Q = -4e$ and excited by an electron ($q = -e$) moving parallel to the chain and at a distance $z_0 = 0.5$ nm away. In each column, the normalized CL emission (upper panel) calculated in linear response (dashed curves) and time-domain simulations (solid curves) is presented for the indicated electron speed above the corresponding time evolution of the induced charge $\rho_l^{\text{ind}} \equiv -2e(\rho_{ll} - \rho_{ll}^{(0)})$ along the chain position x_l (middle panel) and change in eigenstate populations $\rho_j^{\text{ind}} \equiv \rho_{jj} - f_j$ for energies $\hbar\epsilon_j$ around the Fermi energy E_F (lower panel). The pink lines in the middle panel row indicate the free electron trajectory.

plasmon modes of the TI are less saturated than their counterparts in the semiconductor, while the opposite trend is observed for higher-order modes. Results in Fig. 2(b) are in qualitative agreement with the electron-induced nonlinearity presented by doped armchair-edge-terminated graphene nanoislands exhibiting a semiconducting electronic character [20]. The magnitude of the nonlinear response is comparable among the three electronic phases, although the saturation and spectral shifting of CL peaks exhibited by semiconducting and TI chains appear more prominently than the CL enhancement in the metallic chain. While these trends are observed for charged particles traveling parallel to the chain, a perpendicular trajectory results in CL saturation from all three electronic phases (see Supplemental Material [31]).

Trends in the deviation from linear response in the CL and EELS signals corresponding to the lowest-order dipolar plasmon mode exhibit a strong dependence on doping: enhancement or saturation in CL and EELS signals can be controlled by doping, while a critical doping density triggers a transition from redshift to blueshift in the nonlinear response (see Supplemental Material [31]). These results underscore the complexity of the electron-induced nonlinear response in even a simple model system of SSH atomic chains. The significant nonlinearity in lightly doped gapped chains suggests that charged molecules exhibiting a band gap or semiconducting nanoparticles could be ideal platforms to observe nonlinear optical phenomena induced by low-energy electrons.

The electron speed determines the interaction time with optical resonances in the target SSH chain, thus controlling the nonlinear response. Figures 3(a), 3(b) and 3(c) explore induced charge carrier dynamics in metallic, semiconducting,

and TI chains, respectively, each with $N = 100$ sites and $Q = 4$ added electrons, by presenting the lowest-order plasmon CL signal (upper row) produced from the real-space induced charge density ρ_l^{ind} (middle row) and the corresponding occupation factors $\rho_{jj} = \sum_{ll'} a_{jl} a_{jl'} \rho_{ll'}$ (lower row) when free electrons pass a distance 0.5 nm parallel to the chain at speeds of 0.04c (left columns), 0.02c (middle columns) and 0.01c (right columns).

The real-space induced charge oscillations in Fig. 3 (middle row) reveal dynamics leading to nonlinear response in CL and EELS signals: Fast electrons (left panels) pass over the ≈ 14 -nm doped chains within ≈ 2 fs to induce opposite charges at the chain ends that move towards one another at the speed of electrons in the medium (i.e., the Fermi velocity v_F) and interfere, producing complex patterns that are presumably related to the excitation of both plasmon modes and individual electron-hole pairs. Incidentally, the interference effect originating at the ends of the chain is analogous to the transition radiation generated by an electron passing through a medium and propagating at the group velocity of light [33,34]. In contrast, the slow-moving electron (right panels) moves at the same speed as the induced charge, dragging it to the opposite end of the chain to undergo multiple reflections into the available excitation channels. The predicted effect resembles the induced charge waves resulting from the Coulomb drag of an electron moving above a doped graphene sheet [35]. The dynamics described above occur in all phases, but are most prominently seen in the metallic chain due to the abundance of free charge carriers.

In the energy-space basis, the redistribution of population among discrete states in doped atomic chains is most

prominently seen in Fig. 3 (bottom row) around the Fermi energy, where multiple intraband transitions form plasmon resonances. For the metallic chain shown in Fig. 3(a), the fast electron (bottom left panel) weakly perturbs an energetically narrow range of states around E_F that relaxes on a timescale determined by γ^{-1} . In contrast, the slow electron (bottom right panel) displaces electron population over a broad range of energies symmetrically around E_F , increasing the number of intraband transitions that can participate in the collective excitation. The gapped chains considered in Figs. 3(b) and 3(c) also exhibit a strong intraband response in the conduction band, accompanied by signatures of interband excitations. The vanishing density of states in the band gap however leads—particularly in the case of the slow electron—to a broad and asymmetric population displacement in the conduction band that is probed more effectively by the passing electron. Presumably, the nonequilibrium electron distribution resembles that of effectively lower chemical potential and larger electronic temperature, leading to qualitatively similar saturation and spectral shifting of plasmonic resonances as predicted in the transient response of optically pumped graphene nanoribbons [36].

The results of Fig. 3 indicate that the speed of charge carriers in the chain near the Fermi energy in the doped SSH chains determines the threshold speed of a charged particle to trigger a nonlinear response in CL and EELS signals. We confirm this hypothesis by demonstrating that the deviation from linear response diminishes for particle speeds above the Fermi velocity v_F (see Supplemental Material [31]). The Fermi velocity is decreased near the band edge of semiconducting and TI chains, so that they present an enhanced nonlinearity when lightly doped. However, plasmon resonances of gapped chains with fewer free charge carriers couple more weakly with impinging electromagnetic fields and thus appear less prominently than their counterparts in intrinsically metallic chains.

Under grazing incidence, electron-induced optical nonlinearity in the plasmons of doped semiconducting and TI atomic chains manifests as spectral shifts and saturation in CL and EELS signals, while metallic chains exhibit an enhanced plasmonic response. Our results establish the Fermi velocity of free charge carriers as a threshold for triggering electron-induced nonlinear optical effects in plasmon resonances supported by a target nanostructure, suggesting that electronic band engineering plays an important role in such phenomena, where charge carrier doping can further tune v_F and plasmon resonance frequencies. We note that the single-particle formalism employed here does not capture many-body effects, which are important for describing exciton resonances that should be prominent in undoped semiconductors and similarly exhibit an electron-induced nonlinear response. We have considered nonrelativistic free electrons in the no-recoil approximation, which should be reasonable for electrons with energy $\gtrsim 10$ s of eV available in low-energy electron microscopy experiments [37], while heavy charged particles such as ions with even lower speeds could produce dramatic nonlinear effects (see Supplemental Material [31]). These findings guide future experimental efforts to observe electron-induced nonlinear effects in CL and EELS, circumventing use of high-fluence optical pulses to explore nonlinear optical phenomena on the nanoscale.

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