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Floquet theory for the electronic stopping of projectiles in solids

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A theoretical framework for the study of electronic stopping of particle projectiles in crystalline solids is proposed. It neither relies on perturbative or linear-response approximations nor on an ideal metal host. Instead, it exploits the discrete translational invariance in a space-time diagonal for a constant velocity projectile following a trajectory with crystalline periodicity. By means of Floquet theory, (stroboscopically) stationary solutions are characterized, and previous perturbative and jellium models are recovered. The threshold-velocity effect in insulators is analyzed based on quasienergy conservation.

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I. INTRODUCTION

Particles shooting through matter slow down when interacting with nuclei and electrons. The energy loss per unit length to the electrons (nuclei) is called electronic (nuclear) stopping power $S_{e(n)}$. It is of applied interest in various contexts: The effect of radiation in matter is important in nuclear and aerospace industries and for medical applications. From the fundamental side, it represents a quite canonical problem in the field of strongly nonequilibrium electronic systems.

Electronic stopping of ions in solids has been studied for over a century [1–5]. The most popular theoretical paradigm for nonrelativistic velocities is the linear-response theory of Lindhard, of general applicability for any host material [6,7], and accessible to first-principles theory [8]. It assumes, however, a weak effective interaction between the projectile and the solid. A fully nonlinear theory was proposed for the homogeneous electron liquid (jellium) for the low-velocity limit [9], including first-principles calculations [10]. It maps the electronic stopping problem into that of electron scattering by an impurity in jellium when changing to the projectile reference frame. It was later extended to finite v [11–14]. Although a successful theory and fundamental reference for simple metals, its applicability to semiconductors, insulators, transition metals, etc., is qualitative and limited. Extensive work has been performed within these paradigms (see references within Refs. [15,16]).

Explicit simulations of the stopping process are also used in which a projectile moves within a solid in a large simulation box using either time-dependent tight-binding [17–19] or first-principles time-dependent density-functional theory [20–36]. They are computationally expensive but allow the study of materials beyond simple metals and have access [20,27,37] to experimentally observed nontrivial effects, such as the appearance of a threshold velocity [38–42], which also prompted other theoretical investigations [30,43–48]. However, there appears to be no physical theory beyond linear response and jellium addressing the stationary states of stopping for arbitrary crystalline systems, which is the focus of this paper.

II. MODEL

We consider the widely used [5,6,10–14,20,22,24,25,27,29–36] ideal model of a projectile moving at a constant velocity along a rectilinear trajectory. It is a useful model in the characterization of $S_e(v)$, and, experimentally, the slowing down of a keV/MeV projectile by a few eV/Å, is barely noticeable over the nanoscale distances relevant to the processes faced here [20]. It is nonconservative for the electronic system, the energy change compensating the work needed to keep the velocity constant. The following formalism is presented for noninteracting particles, and the projectile is represented by a local scalar potential. The method can be straightforwardly generalized to more realistic situations using time-dependent mean-field or Kohn-Sham methods to include realistic crystals, projectiles, and electron-electron interactions [20–36].

The theory for jellium [9–12] is implicitly built on the fact that the problem of a projectile of constant velocity $\mathbf{v} = v\hat{\mathbf{v}}$ moving in a homogeneous electron liquid, although a time-dependent nonconservative problem, retains a continuous symmetry and related conservation, which neither stems from time nor space homogeneity but rather from invariance along a space-time diagonal. The change to the projectile reference frame aligns this trajectory with the time axis, and the problem becomes energy conservative, whereas still dissipative in the laboratory frame. Consider the projectile in a crystalline solid with a spatial periodicity a along its trajectory. The translational invariance becomes discrete along the same line of space-time: The system is invariant under combined space-time translations $\mathcal{T}^*$: $\mathbf{r} \to \mathbf{r} + na\hat{\mathbf{v}}$, $t \to t + n\tau$ with $n \in \mathcal{Z}$ and $\tau = a/v$. Changing to the projectile

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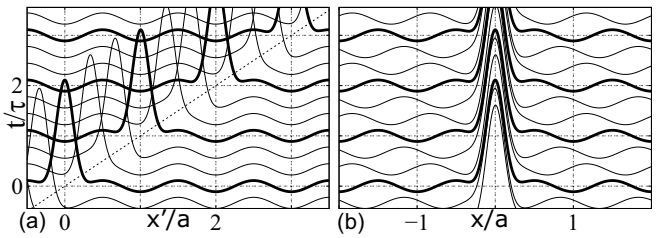

FIG. 1. Evolution of crystalline plus projectile potential in (a) the laboratory reference frame and in (b) the projectile. a is the lattice parameter $\tau = a/v$, and v is the projectile velocity. The curves (potential vs x) are shifted for different times. Thicker lines indicate times separated by τ.

reference frame, the problem becomes purely time periodic with period τ, switching from $\mathcal{T}^*$ invariance to $\mathcal{T}: t \to t + \tau$ invariance (Fig. 1). In this frame, both the host electrons and the crystalline potential move past the projectile with velocity $-\mathbf{v}$. The $\mathcal{T}$ invariance is the main point exploited in this paper as it allows using Floquet theory for time-periodic Hamiltonians [49,50]. A general time-dependent lattice-projectile model Hamiltonian with the $\mathcal{T}^*$ symmetry in the laboratory frame in which the lattice is at rest can be written as

$$H_{\rm lab}(\mathbf{r}', t) = H_0(\mathbf{r}') + V_P(\mathbf{r}', t), \tag{1}$$

where $H_0(\mathbf{r}')$ is the lattice Hamiltonian and $V_P(\mathbf{r}', t) = V_P(\mathbf{r}' - \mathbf{v}t)$ describes a projectile with velocity $\mathbf{v}$. In the reference frame moving with the projectile, $\mathbf{r} = \mathbf{r}' - \mathbf{v}t$ (primed/unprimed indices indicating laboratory/projectile frame, respectively), the Hamiltonian becomes

$$H(\mathbf{r}, t) = H_0(\mathbf{r} + \mathbf{v}t) + V_P(\mathbf{r}), \tag{2}$$

which is time periodic with period τ.

According to Floquet theorem [49,50], for a time-periodic Hamiltonian with period τ, there are time-dependent solutions to the Schrödinger equation of the form

$$\psi_\alpha(\mathbf{r}, t) = e^{-i\varepsilon_\alpha t/\hbar}\phi_\alpha(\mathbf{r}, t), \tag{3}$$

where the Floquet mode satisfies $\phi_\alpha(\mathbf{r}, t) = \phi_\alpha(\mathbf{r}, t + \tau)$. The quasienergy ε_α is conserved and defines solutions up to multiples of $\hbar\omega$, $\omega = 2\pi/\tau$. Quasienergies $\varepsilon_{p\alpha} = \varepsilon_\alpha + p\hbar\omega$ (integer p) are equivalent since $\phi_{p\alpha}(\mathbf{r}, t) = e^{ip\omega t}\phi_\alpha(\mathbf{r}, t)$ is $\mathcal{T}$ periodic. The zeroth mode quasienergy $\varepsilon_{0\alpha} = \varepsilon_\alpha$ is, therefore, defined on any $\hbar\omega$ interval, e.g., $-\hbar\omega/2 < \varepsilon_{0\alpha} \leqslant \hbar\omega/2$, analogous to the first Brillouin zone (BZ). The Floquet modes satisfy

$$\mathcal{H}(\mathbf{r}, t)\phi_{p\alpha}(\mathbf{r}, t) = \varepsilon_{p\alpha}\phi_{p\alpha}(\mathbf{r}, t), \tag{4}$$

with the Floquet Hamiltonian $\mathcal{H}(\mathbf{r}, t) = H(\mathbf{r}, t) - i\hbar\frac{\partial}{\partial t}$.

A first important consequence is that the Floquet modes in the projectile frame define the stationary solutions to the stopping problem in the laboratory frame. In previous work [7,10–12], stationary solutions were either assumed or a direct consequence of key approximations. Their existence and character appear now naturally from Floquet theorem. Stationary now means $\mathcal{T}$ periodic or stroboscopic, i.e., time independent if looking at it at instants $t = t_0 + n\tau$ for $n \in \mathcal{Z}$. It does not mean these are the only expected solutions. In addition to transients related to perturbations, one can also foresee deviations, such as the flapping instability recently proposed [36], which represents the analog of a charge density wave along the $\mathcal{T}^*$-symmetric direction in space-time, a generalization of the time-crystal idea. Moreover, it is known that the effect of dissipation becomes more subtle when going to higher levels of the jellium theory [51,52]; therefore, the character of the Floquet states and the meaning of the quasienergy might have to be revised in future extensions of the present theory. However, this is beyond the scope of this paper.

III. STOPPING FROM BLOCH-FLOQUET SCATTERING THEORY

The stopping problem in the laboratory frame becomes a scattering one for the Floquet modes in the projectile, in analogy with the theory for jellium, replacing energy by quasienergy conservation, and treating time t as an additional degree of freedom at the same level of a spatial coordinate [53,54]. The asymptotic scattering states away from the projectile consist of the Bloch states of the crystal transformed to the projectile frame. Define $\mathcal{H}_0(\mathbf{r}, t) = H_0(\mathbf{r} + \mathbf{v}t) - i\hbar\frac{\partial}{\partial t}$ as the crystal Floquet Hamiltonian, periodic with period τ, whose complete set of eigenmodes $\{\phi_\alpha(\mathbf{r}, t)\}$ are readily extracted from the Bloch states in the laboratory frame $\psi'_{n\mathbf{k}}(\mathbf{r}') = e^{i\mathbf{k}\cdot\mathbf{r}'}u_{n\mathbf{k}}(\mathbf{r}')$ with energy $E_n(\mathbf{k})$ and band index n, which, transformed to the projectile frame, become [55]

$$\psi_{n\mathbf{k}}(\mathbf{r}, t) = u_{n\mathbf{k}}(\mathbf{r} + \mathbf{v}t)e^{i(\mathbf{k} - m\mathbf{v}/\hbar)\cdot\mathbf{r}}e^{-i\varepsilon_n(\mathbf{k})t/\hbar}, \tag{5}$$

where $\varepsilon_n(\mathbf{k}) = E_n(\mathbf{k}) - \hbar\mathbf{k}\cdot\mathbf{v} + mv^2/2$ and m is the electron mass. By comparing with Eq. (3), the quasienergies and Floquet modes are immediately identified as $\varepsilon_\alpha = \varepsilon_n(\mathbf{k})$ and $\phi_\alpha(\mathbf{r}, t) = \phi_{n\mathbf{k}}(\mathbf{r}, t) \equiv u_{n\mathbf{k}}(\mathbf{r} + \mathbf{v}t)e^{i(\mathbf{k} - m\mathbf{v}/\hbar)\cdot\mathbf{r}}$ (Bloch-Floquet modes, henceforth). The Floquet BZ for quasienergy can be chosen to coincide with the BZ for the Bloch vectors: shifting $\mathbf{k}$ by $p\mathbf{G}_0$ [for $p \in \mathcal{Z}$ and $\mathbf{G}_0 = (2\pi/a)\hat{\mathbf{v}}$] shifts the quasienergy by $p\hbar\omega$, $\varepsilon_{pn}(\mathbf{k}) = \varepsilon_n(\mathbf{k} + p\mathbf{G}_0)$.

Consider a Bloch state $\psi_{n\mathbf{k}_i}(\mathbf{r}, t)$ in the moving frame [Eq. (5)]. With the addition of the projectile, which does not break the symmetry $\mathcal{T}$ [the Floquet Hamiltonian is now $\mathcal{H}(\mathbf{r}, t) = \mathcal{H}_0(\mathbf{r}, t) + V_P(\mathbf{r})$], the Floquet mode of the full solution $\Psi^{(\pm)}_{n\mathbf{k}_i} = e^{-i\varepsilon_n(\mathbf{k}_i)t/\hbar}\Phi^{(\pm)}_{n\mathbf{k}_i}(\mathbf{r}, t)$ with the same quasienergy $\varepsilon_n(\mathbf{k}_i)$ can be expressed as an integral equation in the Lippmann-Schwinger spirit with $\xi = (\mathbf{r}, t)$,

$$\Phi^{(\pm)}_{n\mathbf{k}_i}(\xi) = \phi_{n\mathbf{k}_i}(\xi) + \int d\xi' \mathcal{G}^{(\pm)}_0[\varepsilon_n(\mathbf{k}_i)|\xi, \xi']V_P(\mathbf{r}')\Phi^{(\pm)}_{n\mathbf{k}_i}(\xi'), \tag{6}$$

where the $(\pm)$ sign indicates outgoing/incoming boundary conditions, and an averaging over one cycle in t' is implied. $\mathcal{G}^{\pm}_0$ is the propagator for $\mathcal{H}_0$ [53,56],

$$\mathcal{G}^{(\pm)}_0(\varepsilon|\xi, \xi') = \sum_n \sum_p \int_{1BZ} \frac{d\mathbf{k}}{(2\pi)^3} \frac{\phi_{pn\mathbf{k}}(\xi)\phi^*_{pn\mathbf{k}}(\xi')}{\varepsilon - \varepsilon_{pn}(\mathbf{k}) \pm i\eta}, \tag{7}$$

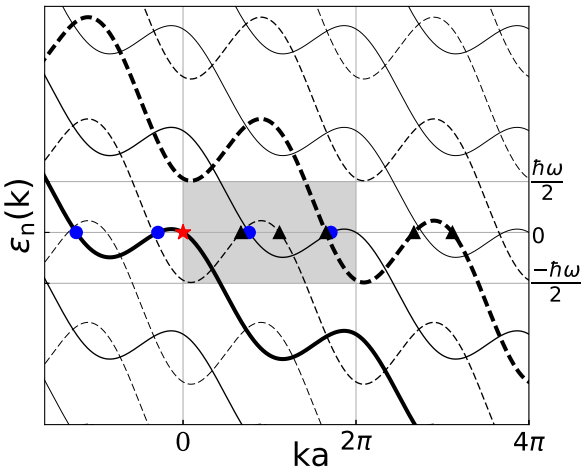

FIG. 2. Bloch-Floquet-mode quasienergies $\varepsilon_n(\mathbf{k}) = E_n(\mathbf{k}) - \hbar\mathbf{k}\cdot\mathbf{v} + \frac{1}{2}mv^2$ of a two-band model along the projectile-periodic direction in reciprocal space, extended zone scheme, and Floquet replicas $\varepsilon_{pn}(\mathbf{k})$ included (thin lines) $v > 0$. The first BZ is highlighted in gray. Quasienergy conserving states are indicated for an incoming mode at $k = 0$ (red star). Blue circles and black triangles label allowed scattering modes in the lower and upper bands, respectively, both in the extended zone scheme (on thick curves) and folded back into the first BZ.

where the sum over higher-order Floquet modes p as defined in the previous paragraph must be included.

The asymptotic behavior of Eq. (6) for an initial Bloch-Floquet wave with outgoing boundary conditions is

$$\Phi^{(+)}_{n\mathbf{k}_i}(\mathbf{r},t) \sim \phi_{n\mathbf{k}_i}(\mathbf{r},t) + \sum_{m,k_f} A_{n\mathbf{k}_i,mk_f\hat{\mathbf{r}}} \frac{e^{i(k_f\hat{\mathbf{r}} - m\mathbf{v}/\hbar)\cdot\mathbf{r}}}{r} u_{mk_f\hat{\mathbf{r}}}(\mathbf{r},t), \tag{8}$$

where the scattering amplitudes are defined through a generalized inner product [49] $\langle\langle f|g\rangle\rangle = \frac{1}{\tau}\int_0^\tau dt \int d\mathbf{r}\, f^*(\mathbf{r},t)g(\mathbf{r},t)$ as

$$A_{n\mathbf{k}_i,mk_f\hat{\mathbf{r}}} = -\frac{k_f}{2\pi\hbar v_g^m(k_f\hat{\mathbf{r}})} \left\langle\!\left\langle \phi_{mk_f\hat{\mathbf{r}}}(t)\middle|V_P\middle|\Phi^{(+)}_{n\mathbf{k}_i}(t)\right\rangle\!\right\rangle, \tag{9}$$

with the group velocities of the outgoing states $v_m^g(k_f\hat{\mathbf{r}}) = \frac{1}{\hbar}\frac{\partial}{\partial k}[\varepsilon_m(k\hat{\mathbf{r}})]|_{k=k_f} > 0$. The values of m and $\mathbf{k}_f$ for the outgoing modes are determined by quasienergy conservation $\varepsilon_n(\mathbf{k}_i) = \varepsilon_m(\mathbf{k}_f)$, which has, in general, multiple solutions (Fig. 2). It can also be written, using the definition for $\varepsilon_n(\mathbf{k})$ [below Eq. (5)] as

$$E_m(\mathbf{k}_f) - E_n(\mathbf{k}_i) = \hbar(\mathbf{k}_f - \mathbf{k}_i)\cdot\mathbf{v}\,. \tag{10}$$

This expression, appearing naturally from quasienergy conservation, coincides with that obtained from energy and momentum conservation in a binary collision of an electron with a projectile of mass $M_P \to \infty$ [27] and, in perturbation theory [57], with the distinction that $\mathbf{k}$ values must be considered in the Bloch-Floquet extended zone/repeated band scheme (see Fig. 2).

For the single-particle scattering state of Eq. (8), the energy transfer rate (ETR) is derived from the energy flux difference between incident and scattered modes. Using conservation of

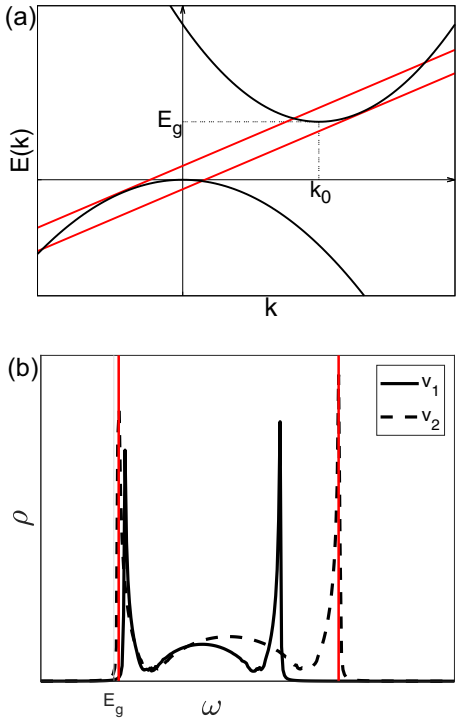

FIG. 3. (a) Model parabolic bands with indirect band gap in one dimension (1D) for illustration. Red lines delimit possible electron-hole pair transitions compatible with Eq. (10) with projectile velocity v defining their slope. (b) JDOS $\rho(\omega, v)$ vs excitation energy ω for velocities $v_2 > v_1 > v_{\rm th}$ for the same model. Red vertical lines highlight van Hove singularities.

probability flux, the ETR can be written as

$$\dot{E}_{ni} = \rho_i \frac{2\pi}{\hbar}\int \frac{d\mathbf{k_f}}{(2\pi)^3}\Delta E_{mn,fi}\left|\left\langle\!\left\langle \phi_{m\mathbf{k}_f}\middle|V_P\middle|\Phi^{(+)}_{n\mathbf{k}_i}\right\rangle\!\right\rangle\right|^2 \times \delta(\Delta E_{mn,fi} - \mathbf{v}\cdot\hbar\,\Delta\mathbf{k}_{fi}), \tag{11}$$

where ρ_i is the density of the incoming state, $\Delta E_{mn,fi} = E_m(\mathbf{k}_f) - E_n(\mathbf{k}_i)$, and $\Delta\mathbf{k}_{fi} = \mathbf{k}_f - \mathbf{k}_i$ with the Dirac-δ imposing quasienergy conservation. Electronic stopping can be defined as $S_e = \dot{E}/v$, $\dot{E}$ being the total ETR. At temperature $T = 0$, assuming occupied bands n and unoccupied bands m and integrating separately over initial and final modes,

$$S_e(\mathbf{v}) = \frac{1}{v}\sum_{nm}\frac{2\pi}{\hbar}\int\frac{d\mathbf{k}_i}{(2\pi)^3}\int\frac{d\mathbf{k}_f}{(2\pi)^3}\Delta E_{mn,fi} \times \left|\left\langle\!\left\langle \phi_{m\mathbf{k_f}}\middle|V_P\middle|\Phi^{(+)}_{n\mathbf{k_i}}\right\rangle\!\right\rangle\right|^2 \delta(\Delta E_{mn,fi} - \mathbf{v}\cdot\hbar\,\Delta\mathbf{k}_{fi}). \tag{12}$$

To extend this to $T \neq 0$ and partially filled bands, the relevant occupation numbers can be introduced. The jellium theory [9–12] is recovered from Eq. (11) for the ETR and from Eq. (12) for stopping. Alternatively, if the projectile is treated as a weak perturbation, the equivalent of a first Born approximation for Floquet scattering can be used [58]: Substituting $|\Phi^{(+)}_{n\mathbf{k}_i}\rangle$ by $|\phi_{n\mathbf{k}_i}\rangle$ and assuming a smooth projectile $V_P(\mathbf{r})$, the matrix element [Eq. (9)] becomes

$$\left\langle\!\left\langle \phi_{m\mathbf{k}_f}\middle|V_P\middle|\phi_{n\mathbf{k}_i}\right\rangle\!\right\rangle \propto \tilde{V}_P(\Delta\mathbf{k}), \tag{13}$$

where $\Delta\mathbf{k} = |\mathbf{k}_f - \mathbf{k}_i|$ and $\tilde{V}_P$ indicates the Fourier transform of $V_P(\mathbf{r})$ thereby recovering perturbation theory results (see, e.g., Ref. [57]).

IV. THRESHOLD VELOCITY FOR INSULATORS

The low-v limit for the stopping of ions in a gapped material, which proved to be quite controversial in experiments [39,40,59], is now analyzed. From the theory side, it was studied both with linear-response approaches and adapting the jellium nonlinear theory by introducing phenomenological restrictions to the transport cross section (see, e.g., Ref. [48]). Our theory allows addressing the full nonlinear insulating problem. The constant-velocity assumption retains validity since, below a velocity threshold, the projectile slowing down is negligible even for keV projectiles or below. The effect of the crystal on v remains typically small at that scale, but note that the theory is valid for a periodically varying velocity as long as the average is constant. Consider a model insulator (at the independent particle level) with parabolic energy bands around the gap and isotropic effective masses for electrons and holes with an indirect band-gap E_g, the bottom of the conduction band displaced by $\mathbf{k}_0$ from the top of the valence band, and a projectile with velocity $\mathbf{v} = v\hat{\mathbf{k}}_0$.

This simple model does not include plasmonic contributions to stopping or quantitative predictions for the electron-hole contributions, which demand including electron-electron interaction within Eq. (12). The theory is ready for including known techniques for the quantitative description of electron correlations as needed. At this stage, however, the simple model already offers good insights into what to expect for the electron-hole excitation contribution in the nonlinear gapped case. A joint density of states (JDOS) can be defined in analogy with optical transitions [60],

$$\rho(\omega, v) = \sum_{nm} \int \frac{d\mathbf{k}_i}{(2\pi)^3} \int \frac{d\mathbf{k}_f}{(2\pi)^3} \delta[\varepsilon_m(\mathbf{k}_f) - \varepsilon_n(\mathbf{k}_i)] \times \delta(\Delta E_{mn,fi} - \hbar\omega), \tag{14}$$

offering interesting insights—see Fig. 3 for 1D, illustrative of the behavior in any dimensions. For the parabolic model in the figure, no stopping is allowed below a threshold velocity v_{th}. An integration of the JDOS gives $S_e(v) = f(v - v_{\text{th}})$, $v > v_{\text{th}}$. When $v \gtrsim v_{\text{th}}$, $f(v) \propto v^m$, where m depends on dimension [$m = 1$ in 1D, $m = 2$ in three dimensions (3D)]. For an actual insulator, however, the threshold behavior is less clean. In fact, the adiabatic limit $v \to 0$ is quite nontrivial as illustrated in Fig. 4: By quasienergy conservation [Eq. (10)] transitions are allowed for arbitrarily small v even for gapped solids. This is shown in the figure using the repeated zone scheme where the lines of allowed transitions decrease in slope with decreasing v. Importantly, this picture is general and independent of perturbation theory [46,57].

The $S_e(v)$ curve, which is given by the sum over all the contributions for stopping from each parabolic replica, is characterized by a series of onset velocities or partial thresholds $v_{\text{th}}^{(p)}$ for $p \in \mathcal{Z}_{\geqslant 0}$ defined by

$$E_g = \frac{1}{2}(m_e + m_h)\left(v_{\text{th}}^{(p)}\right)^2 + \hbar\left(k_0 + \frac{2\pi}{a}p\right)v_{\text{th}}^{(p)}. \tag{15}$$

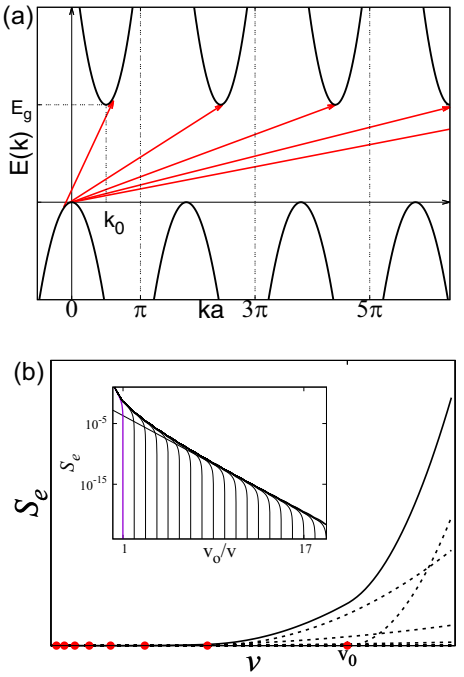

FIG. 4. (a) Partial threshold velocities (slopes of red lines $v_{\text{th}}^{(p)}$) for replicas of parabolic bands in the extended zone scheme, corresponding to shifted Floquet modes, cf. Fig. 2. (b) Effective threshold behavior for S_e vs v in the small v limit for a 3D indirect gap model with $\gamma_l \propto e^{-\alpha l}$. Red dots: $v_{\text{th}}^{(p)}$, v_0 is the threshold velocity for transitions within the first BZ. The inset: S_e (logarithmic scale) vs $1/v$ highlights the quick decay of stopping as $v \to 0$.

In the low-v limit (large p), $v_{\text{th}}^{(p)} \sim \frac{E_g}{2\pi/a}\frac{1}{p}$. In this limit, assuming a decaying scattering rate γ_l for transitions to the lth replica, $S_e(v)$ can be accurately approximated by a semi-infinite sum over the higher-order replica contributions, each term as discussed above for the single parabola case. For $v \approx v_{\text{th}}^{(p)}$, it becomes

$$S_e(v_{\text{th}}^{(p)}) \approx \sum_{l=p}^{\infty} \gamma_l f\left(v_{\text{th}}^{(p)} - v_{\text{th}}^{(l)}\right). \tag{16}$$

For a slow algebraic decay $\gamma_l \sim l^{-\mu}$,

$$S_e(v_{\text{th}}^{(p)}) \approx S_0 \sum_{l=p}^{\infty} \left(\frac{1}{p} - \frac{1}{l}\right)^m \frac{1}{l^\mu}, \tag{17}$$

giving the low-v behavior $S_e(v) \sim v^{m+\mu-1}$. A quicker decay $\gamma_l \sim e^{-\alpha l^\lambda}$ implies $S_e(v) \sim e^{-(v^*/v)^\lambda}$, which would lead to an observed "hard" threshold. In Fig. 4(b), this behavior for the 3D parabolic model is shown, assuming an exponential decay for γ_l, highlighting how the threshold emerges from the theory. The behavior predicted, here, below the primary threshold v_0 has not been resolved by experiments so far (see Fig. 2 in Ref. [39] where the reported error bar represents half of the vertical scale in Fig. 4).

To summarize, the presented theory provides a natural framework for describing the stroboscopic stationary states arising in electronic stopping processes as well as the reference states for possible instabilities along the space-time symmetric direction, analogous to charge-density waves in

space or time crystals in time. The theory constitutes the basis for future first-principles methods directly addressing stationary solutions for electronic stopping processes. Previous perturbative [57,58] and nonlinear jellium [10–12] theories are recovered in the appropriate limits. Floquet quasienergy conservation has allowed the characterization of nontrivial velocity thresholds in insulators.

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[1] W. H. Bragg and R. Kleeman, London, Edinburgh, Dublin Philos. Mag. **10**, 318 (1905).
[2] N. Bohr, Lonon, Edinburgh, Dublin Philos. Mag. **25**, 10 (1913).
[3] H. Bethe, Ann. Phys. (NY) **397**, 325 (1930).
[4] F. Bloch, Ann. Phys. (NY) **408**, 285 (1933).
[5] E. Fermi and E. Teller, Phys. Rev. **72**, 399 (1947).
[6] J. Lindhard, Kgl. Danske Videnskab. Selskab Mat.-Fys. Medd. **28** (1954).
[7] J. Lindhard, M. Scharff, and H. Schiøtt, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **33** (1963).
[8] A. A. Shukri, F. Bruneval, and L. Reining, Phys. Rev. B **93**, 035128 (2016).
[9] T. L. Ferrell and R. H. Ritchie, Phys. Rev. B **16**, 115 (1977).
[10] P. Echenique, R. Nieminen, and R. Ritchie, Solid State Commun. **37**, 779 (1981).
[11] K. Schönhammer, Phys. Rev. B **37**, 7735 (1988).
[12] L. Bönig and K. Schönhammer, Phys. Rev. B **39**, 7413 (1989).
[13] E. Zaremba, A. Arnau, and P. Echenique, Nucl. Instrum. Methods Phys. Res., Sec. B **96**, 619 (1995).
[14] A. F. Lifschitz and N. R. Arista, Phys. Rev. A **57**, 200 (1998).
[15] P. Sigmund, *Particle Penetration and Radiation Effects* (Springer, Berlin, 2006).
[16] P. Sigmund, *Particle Penetration and Radiation Effects Volume 2* (Springer, Berlin, 2014).
[17] D. R. Mason, J. le Page, C. P. Race, W. M. C. Foulkes, M. W. Finnis, and A. P. Sutton, J. Phys.: Condens. Matter **19**, 436209 (2007).
[18] C. P. Race, D. R. Mason, M. W. Finnis, W. M. C. Foulkes, A. P. Horsfield, and A. P. Sutton, Rep. Prog. Phys. **73**, 116501 (2010).
[19] C. P. Race, D. R. Mason, M. H. F. Foo, W. M. C. Foulkes, A. P. Horsfield, and A. P. Sutton, J. Phys.: Condens. Matter **25**, 125501 (2013).
[20] J. M. Pruneda, D. Sánchez-Portal, A. Arnau, J. I. Juaristi, and E. Artacho, Phys. Rev. Lett. **99**, 235501 (2007).
[21] A. V. Krasheninnikov, Y. Miyamoto, and D. Tománek, Phys. Rev. Lett. **99**, 016104 (2007).
[22] M. Quijada, A. G. Borisov, I. Nagy, R. Díez Muiño, and P. M. Echenique, Phys. Rev. A **75**, 042902 (2007).
[23] R. Hatcher, M. Beck, A. Tackett, and S. T. Pantelides, Phys. Rev. Lett. **100**, 103201 (2008).
[24] A. A. Correa, J. Kohanoff, E. Artacho, D. Sánchez-Portal, and A. Caro, Phys. Rev. Lett. **108**, 213201 (2012).
[25] M. A. Zeb, J. Kohanoff, D. Sánchez-Portal, A. Arnau, J. I. Juaristi, and E. Artacho, Phys. Rev. Lett. **108**, 225504 (2012).
[26] A. Ojanperä, A. V. Krasheninnikov, and M. Puska, Phys. Rev. B **89**, 035120 (2014).
[27] R. Ullah, F. Corsetti, D. Sánchez-Portal, and E. Artacho, Phys. Rev. B **91**, 125203 (2015).
[28] Z. Wang, S.-S. Li, and L.-W. Wang, Phys. Rev. Lett. **114**, 063004 (2015).
[29] A. Schleife, Y. Kanai, and A. A. Correa, Phys. Rev. B **91**, 014306 (2015).
[30] A. Lim, W. M. C. Foulkes, A. P. Horsfield, D. R. Mason, A. Schleife, E. W. Draeger, and A. A. Correa, Phys. Rev. Lett. **116**, 043201 (2016).
[31] E. E. Quashie, B. C. Saha, and A. A. Correa, Phys. Rev. B **94**, 155403 (2016).
[32] K. G. Reeves, Y. Yao, and Y. Kanai, Phys. Rev. B **94**, 041108(R) (2016).
[33] C.-K. Li, F. Wang, B. Liao, X.-P. OuYang, and F.-S. Zhang, Phys. Rev. B **96**, 094301 (2017).
[34] D. C. Yost, Y. Yao, and Y. Kanai, Phys. Rev. B **96**, 115134 (2017).
[35] G. Bi, J. Kang, and L.-W. Wang, Phys. Chem. Chem. Phys. **19**, 9053 (2017).
[36] R. Ullah, E. Artacho, and A. A. Correa, Phys. Rev. Lett. **121**, 116401 (2018).
[37] R. Cabrera-Trujillo, J. R. Sabin, Y. Öhrn, and E. Deumens, Phys. Rev. Lett. **84**, 5300 (2000).
[38] M. D. Brown and C. D. Moak, Phys. Rev. B **6**, 90 (1972).
[39] S. N. Markin, D. Primetzhofer, and P. Bauer, Phys. Rev. Lett. **103**, 113201 (2009).
[40] M. Draxler, S. P. Chenakin, S. N. Markin, and P. Bauer, Phys. Rev. Lett. **95**, 113201 (2005).
[41] C. Celedón, E. A. Sánchez, M. S. Moreno, N. R. Arista, J. D. Uribe, M. Mery, J. E. Valdés, and P. Vargas, Phys. Rev. A **88**, 012903 (2013).
[42] J. E. Valdés, P. Vargas, and V. A. Esaulov, Radiat. Eff. Defects Solids **171**, 60 (2016).
[43] J. J. Dorado and F. Flores, Phys. Rev. A **47**, 3062 (1993).
[44] I. Nagy, C. Eppacher, and D. Semrad, Phys. Rev. B **62**, 5270 (2000).
[45] I. Nagy, I. Aldazabal, and M. L. Glasser, J. Phys. B: At., Mol. Opt. Phys. **45**, 095701 (2012).
[46] A. P. Horsfield, A. Lim, W. M. C. Foulkes, and A. A. Correa, Phys. Rev. B **93**, 245106 (2016).
[47] A. Lifschitz and N. Arista, Nucl. Instrum. Methods Phys. Res., Sec. B **316**, 245 (2013).
[48] C. D. Archubi and N. R. Arista, Eur. Phys. J. B **89**, 86 (2016).
[49] M. Grifoni and P. Hänggi, Phys. Rep. **304**, 229 (1998).
[50] J. H. Shirley, Phys. Rev. **138**, B979 (1965).
[51] V. U. Nazarov, J. M. Pitarke, C. S. Kim, and Y. Takada, Phys. Rev. B **71**, 121106(R) (2005).
[52] V. U. Nazarov, J. M. Pitarke, Y. Takada, G. Vignale, and Y.-C. Chang, Phys. Rev. B **76**, 205103 (2007).

[53] D. F. Martinez, J. Phys. A **36**, 9827 (2003).
[54] H. Sambe, Phys. Rev. A **7**, 2203 (1973).
[55] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics: Non-relativistic Theory*, 3rd Edition (Pergamon, New York, 1977).
[56] E. N. Economou, *Green's Functions in Quantum Physics*, Springer Series in Solid State Science (Springer, Berlin, 1983), Vol. 7.
[57] E. Artacho, J. Phys.: Condens. Matter **19**, 275211 (2007).
[58] T. Bilitewski and N. R. Cooper, Phys. Rev. A **91**, 033601 (2015).
[59] S. P. Møller, A. Csete, T. Ichioka, H. Knudsen, U. I. Uggerhøj, and H. H. Andersen, Phys. Rev. Lett. **93**, 042502 (2004).
[60] O. Madelung, *Introduction to Solid-State Theory* (Springer, Berlin, 1978).