Stochastic treatment of nonequilibrium ion stopping in solids

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We study the energy loss of fast, hydrogenlike ions in thin solid foils, in the regime prior to the establishment of the ion-charge equilibrium. The projectile-charge evolution is described by a nonstationary, continuous-time Markov process, while the target response is described by a time-dependent dielectric-response formalism. We first derive the projectile self-energy in the presence of charge exchange, which is used to determine the bound-electron density in a self-consistent manner, by minimizing the total projectile energy in an adiabatic approximation. An expression for the ion energy-loss distribution is then used to derive the average value of the stopping power as a function of the traversal time in the foil, taking into account the projectile screening by the bound electron. The results of calculations for He ions in Al foils show significant coherence effects on the energy losses in the pre-equilibrium regime, which are interpreted by the overlap between the time delay in the target response and the characteristic time scale for the charge-changing collisions of the projectile with the target atoms.

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I. INTRODUCTION

The charge state of fast ions traversing solid targets continues to present a challenging research problem in the area of particle interactions with matter [1,2]. Increasing attention has been payed in recent years to experimental studies of energy losses of fast, hydrogenlike ions, passing through solid films with thicknesses comparable to the characteristic length for the projectile charge-state equilibration in such targets [3–5]. This, so-called pre-equilibrium, regime of measurement of ion interactions with solids provides, on the one hand, access to studying the role played by electrons bound to the projectile in ion stopping at fixed charge states [3,4] and, on the other hand, enables experimental determination of the electron capture and loss cross sections, along with the accompanying energy losses, for projectile scattering on target atoms [3,5]. While such powerful experimental techniques provide much needed insight into the problem of ion-charge states during the penetration through solids, there remain several conceptual questions to be addressed in relation to the interpretation of data obtained from very thin targets [6].

On the theoretical side, the role of projectile-charge states in energy losses of light ions in the equilibrium regime has been successfully described by means of the dielectric-response formalism [7,8]. In addition, a comprehensive statistical theory has been developed for the nonequilibrium ion energy-loss spectra in gaseous and solid targets in the presence of charge-changing collisions with target atoms or molecules [9]. Although these two approaches are based on two different points of view, one emphasizing the collective nature of the solid target response [7,8] and the other concentrating on the atomistic picture of target [9], some of their results share a common feature. Namely, the rate of energy deposition in such theories [7–9] is represented by an *incoherent* superposition of contributions from various projectile states, weighted by the *instantaneous* projectile-charge-state distribution.

However, one expects that the overlap among the characteristic time scales for charge-changing processes, undertaken by the projectile, and the typical response time for collective excitations of the quasi-free-electron gas in a solid, would give rise to important *coherent* effects, such that the energy deposition rate in the target would retain some *memory* of the charge-state evolution of the projectile. Such a claim may be corroborated by a very recent experimental study of the femtosecond kinetics of a nonequilibrium electron-hole plasma in GaAs, which has shown that the onset of collective response, such as Coulomb screening and plasmon scattering, exhibits a distinct time delay of the order of the inverse plasma frequency ω_p [10]. Moreover, a closely related phenomenon of transient excitons in metals has been recently interpreted by means of a time-dependent dielectric-response model [11], emphasizing that a finite time is needed for the target to build up a polarization cloud in a reaction to a sudden external perturbation. In our context, considering the finite response time of the electron gas, the charge-changing collisions of the projectile with target atoms may be viewed as a time sequence of abrupt changes in the projectile-charge density, which are separated by randomly distributed time intervals on the sub-femtosecond scale, so that one expects that each such event would be followed by a relatively short period of rather incomplete screening of the projectile by the valence-electron gas in the target. Depend-

ing on the relevant time scales, such recurring periods of incomplete screening of the projectile may partially overlap one another, thus, affecting both the mechanisms of energy deposition in the target and the mechanisms governing the electron binding to the projectile. Considering the transience and the magnitude of the charge-state changes in the earliest stages of ion penetration through a solid, one expects that the pre-equilibrium regime of ion stopping should be particularly affected by such manifestations of the incomplete screening.

In the present paper, we use stochastic methods to describe the coherence effects on the pre-equilibrium ion stopping, in order to reveal what kind of results may be expected for the ion energy losses after very short dwell times in a solid, as well as to elucidate the range of time scales validating the standard picture of ion stopping as an incoherent superposition over the instantaneous charge-state distribution [7–9]. A natural theoretical framework for describing the time-delay effects in the collective excitations of the target electrons is provided by the time-dependent dielectric-response formalism [11,14–16]. We consider hydrogenlike projectiles moving at the medium-to-high speeds, commensurate with the pre-equilibrium experimental regimes [3–5] where the projectile beam consists of predominantly fully stripped ions and the projectiles with only one bound electron. Consequently, the time sequence of the charge-changing events, undertaken by the projectile in collisions with target atoms, is modeled in the pre-equilibrium regime by a nonstationary, continuous-time dichotomic Markov process, which may be easily generalized for projectiles where more than two charge states are relevant by using multivariate Markov processes [17,18]. We generalize here the earlier result for the nonstationary time-correlation function for pointlike projectiles [19], and derive the correlation function for a full time- and space- dependent projectile-charge density. In that context, the bound-electron density is obtained from a self-consistent variational treatment, by minimizing the total energy of the ion [8,13], where the interaction with the target is described by means of the self-energy approach [20].

Calculations are performed with the parameters appropriate for helium projectiles passing through an aluminum target at speeds v in excess of a few Bohr speeds, for which reliable data on the electron capture and loss cross sections $\sigma_{C,L}$ are available, showing that the fraction of neutral projectiles in the beam is negligible [7,12,13]. Since the coherence effects on the ion energy loss and the self-energy are associated with the valence-electron excitation modes, we use simple models for the dielectric function to describe the target response [22] and, in doing so, we neglect the atomistic processes, such as the core-electron excitations and the energy loss in charge-changing collisions with the target atoms. While the contribution of these processes to the total-energy losses of the helium beams in the indicated range of speeds is about ten percent [7,8], we note that the atomistic processes are not likely to exhibit significant coherence effects since they depend on the instantaneous projectile-charge state and are, therefore, not of immediate interest here. We also neglect the role of surface response modes [21], setting a lower limit to the thickness of solid films which may be described by the present model. Consequently, the results obtained here will have only qualitative and semi-quantitative utility in elucidating the importance of the coherence effects on ion stopping in the pre-equilibrium regime, while a fully quantitative treatment would require a generalization which combines the present approach with the standard incoherent theories, where a full account of the atomistic processes and the surface effects could be taken [7,9,21].

In Sec. II, we provide a stochastic description of the projectile-charge density, evaluate the time-dependent self-energy of the projectile, and use it to determine the bound-electron density. In Sec. III, we evaluate the expected pre-equilibrium energy loss and analyze the coherence effects by means of the time-dependent generalization of the concept of the so-called effective stopping charge [2,23]. Concluding remarks are given in Sec. IV. Atomic units are used throughout.

II. PROJECTILE-CHARGE DENSITY

Assume that a fast, hydrogenlike projectile with the atomic number Z traverses a thin foil at a (constant) velocity $\mathbf{v}$, such that the temporal evolution of its charge state $\mathcal{Q}(t)$ is a Markov process which takes two discrete values, $Q_1=Z-1$ and $Q_2=Z$, corresponding to the projectile with one bound electron and to the completely stripped ion, respectively [18]. The characteristic time scale for such a process is determined by the electron capture and loss rates $\Gamma_{C,L}=N_{at}v\sigma_{C,L}$, connecting the two charge states, where N_{at} is the atomic density of the target. Consequently, the full space- and time-dependent projectile-charge density, $\mathcal{D}(\mathbf{r},t)$, defined in the moving frame of reference attached to the projectile, appears to be a random function of time as well, governed by the same statistics as the process $\mathcal{Q}(t)$, such that $\mathcal{Q}(t)\equiv\int d^3\mathbf{r}\mathcal{D}(\mathbf{r},t)$. Therefore, the function $\mathcal{D}(\mathbf{r},t)$ assumes the realizations $\rho_1(\mathbf{r})=Z\delta(\mathbf{r})-\rho_b(\mathbf{r})$ and $\rho_2(\mathbf{r})=Z\delta(\mathbf{r})$ when the projectile-charge state $\mathcal{Q}(t)$ takes the values Q_1 and Q_2, respectively. Here, the normalized bound-electron density is given by $\rho_b(\mathbf{r})=|\psi(\mathbf{r})|^2$, where $\psi(\mathbf{r})$ is a variationally optimized hydrogenic wave function.

Assuming that the electron gas in the solid is described by means of a (retarded) inverse dielectric function $\epsilon^{-1}(\mathbf{r}-\mathbf{r}',t-t')$ [11], the induced potential in the target, $\tilde{V}_{ind}(\mathbf{r},t)$, and the potential of the moving ion, $\tilde{V}_{ext}(\mathbf{r},t)$, are related via $\tilde{V}_{ind}(\mathbf{r},t)=\int d^3\mathbf{r}'\int dt'[\epsilon^{-1}(\mathbf{r}-\mathbf{r}',t-t')-\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')]\tilde{V}_{ext}(\mathbf{r}',t')$, where the tildes signify that both potentials are defined in the target frame of reference. With the projectile-charge density in the target frame being given by the translational shift of the density in the projectile frame, $\tilde{\mathcal{D}}(\mathbf{r},t)=\mathcal{D}(\mathbf{r}-\mathbf{v}t,t)$, one can deduce the induced potential in the projectile frame, $V_{ind}(\mathbf{r},t)$, from the analogous relation, $\tilde{V}_{ind}(\mathbf{r},t)=V_{ind}(\mathbf{r}-\mathbf{v}t,t)$. On introducing the Fourier transform with respect to the space coordinates, we obtain the expected value of the instantaneous self-energy of the projectile at time t, $\langle U(t)\rangle$, as follows:

$$\langle U(t)\rangle \equiv \left\langle \frac{1}{2}\int d^3\mathbf{r}\mathcal{D}(\mathbf{r},t)V_{ind}(\mathbf{r},t)\right\rangle$$
$$= \frac{1}{2}\int \frac{d^3\mathbf{k}}{(2\pi)^3}\frac{4\pi}{k^2}\int_0^t dt'[\epsilon^{-1}(\mathbf{k},t-t') - \delta(t-t')]e^{i\mathbf{k}\cdot\mathbf{v}(t-t')}\langle \mathcal{D}(\mathbf{k},t)\mathcal{D}(\mathbf{k},t')\rangle. \quad (1)$$

It should be noted here that the space Fourier transform of the charge density in the projectile frame, $\mathcal{D}(\mathbf{k},t)$, is also a random function of time, governed by the same statistics as $\mathcal{D}(\mathbf{r},t)$ and $\mathcal{Q}(t)$. Consequently, the expected value in Eq. (1) is obtained by means of the ensemble average over all possible realizations of the process $\mathcal{Q}(t)$, indicated by the symbol $\langle\cdots\rangle$.

Equation (1) indicates that the projectile self-energy at time t is determined by the history of the projectile-charge-density evolution over all preceding times t', starting from the entrance in the target (that is, $t\geqslant t'\geqslant 0$), by means of the time autocorrelation function $\langle\mathcal{D}(\mathbf{k},t)\mathcal{D}(\mathbf{k},t')\rangle$. In order to evaluate this function, we make use of the conditional probability for the process $\mathcal{Q}(t)$ to have the value Q_j at the time t, if its value was Q_l at the time t_0 $(t\geqslant t_0\geqslant 0)$, which is given by $\Pi(Q_j,t|Q_l,t_0)=\phi_j+(\delta_{jl}-\phi_j)\exp[-\Gamma(t-t_0)]$ [18]. Here, ϕ_j is the equilibrium fraction of the charge state Q_j, δ_{jl} is the Kronecker delta, while $\Gamma=\Gamma_C+\Gamma_L$ is the total charge transfer rate. Assuming that the initial charge state at time 0 is Q_0 (equal to either Q_1 or Q_2), we obtain [18,19]

$$\langle\mathcal{D}(\mathbf{k},t)\mathcal{D}(\mathbf{k},t')\rangle \equiv \sum_{j=1}^{2}\sum_{l=1}^{2}\rho_j(\mathbf{k})\rho_l(\mathbf{k})\Pi(Q_j,t|Q_l,t')\times\Pi(Q_l,t'|Q_0,0)$$
$$= \rho_s^2+\rho_c^2e^{-\Gamma(t-t')}+(\rho_0\rho_s-\rho_s^2)e^{-\Gamma t'} + (\rho_0^2-\rho_0\rho_s-\rho_c^2)e^{-\Gamma t}. \quad (2)$$

Here, ρ_s stands for the space Fourier transform of the (equilibrium) projectile-charge density in the stationary regime, given by $\rho_s(\mathbf{k})=\rho_1(\mathbf{k})\phi_1+\rho_2(\mathbf{k})\phi_2$, where $\phi_1=\Gamma_C/\Gamma$ and $\phi_2=\Gamma_L/\Gamma$, while the temporal correlations are described by the charge density $\rho_c(\mathbf{k})=|\rho_2(\mathbf{k})-\rho_1(\mathbf{k})|\sqrt{\phi_1\phi_2}$. In Eq. (2), ρ_0 stands for the space Fourier transform of the initial ion-charge density [either $\rho_1(\mathbf{k})$ or $\rho_2(\mathbf{k})$].

Since the time delay in the collective response of the target electron gas is typically accompanied by the damped oscillations at the plasma frequency ω_p [10,11], it is also of interest to evaluate the time average of the projectile self-energy over the traversal time T through the target, where the plasma oscillations would be washed out, as follows:

$$\overline{\langle U(T)\rangle} \equiv \frac{1}{T}\int_0^T dt\langle U(t)\rangle$$
$$= \frac{1}{2}\int \frac{d^3\mathbf{k}}{(2\pi)^3}\frac{4\pi}{k^2}\int_0^\infty d\omega\,\mathrm{Re}[\epsilon^{-1}(\mathbf{k},\omega)-1]\times F(\mathbf{k},\omega-\mathbf{k}\cdot\mathbf{v};T). \quad (3)$$

Here, $F(\mathbf{k},\omega;T)$ is the spectral function [18] of the nonstationary process $\mathcal{D}(\mathbf{k},t)$, given by

$$F(\mathbf{k},\omega;T)=\frac{1}{\pi}\int_0^T d\tau\cos(\omega\tau)\frac{1}{T}\int_0^{T-\tau}dt\langle\mathcal{D}(\mathbf{k},t+\tau)\mathcal{D}(\mathbf{k},t)\rangle, \quad (4)$$

which is easily evaluated in an analytical form by using Eq. (2), but is too long to be reproduced here. Note, however, that the stationary-regime result is achieved by taking the limit $T\to\infty$ in Eq. (4), in which case we obtain the familiar result [16], $F_s(\mathbf{k},\omega)=\rho_s^2(\mathbf{k})\delta(\omega)+\rho_c^2(\mathbf{k})(\Gamma/\pi)/(\omega^2+\Gamma^2)$. It is also interesting to note that the spectral function approaches the stationary regime in such a way that the prestationary effects decay as the inverse of the traversal time T, viz.,

$$F(\mathbf{k},\omega;T)\sim F_s(\mathbf{k},\omega)+\frac{1}{\Gamma T}\left[(\rho_0\rho_s-\rho_s^2)\delta(\omega) + \frac{\Gamma/\pi}{\omega^2+\Gamma^2}\left(\rho_0^2-\rho_0\rho_s-\rho_c^2\frac{2\Gamma^2}{\omega^2+\Gamma^2}\right)\right]. \quad (5)$$

Let us finally note that Eq. (3), when taken in the stationary regime with the neglect of charge-changing events by setting $\phi_1=1$ and $\phi_2=0$, yields the same expression for the self-energy of a projectile with a bound electron, as the one derived by the many-body techniques [20].

In order to determine the charge density of the bound electron, $\rho_b(\mathbf{r})=|\psi(\mathbf{r})|^2$, in a self-consistent manner, we adopt the procedure where the total projectile energy $\mathcal{E}_{tot}=\langle\psi|-\nabla^2/2-Z/r|\psi\rangle+V_{int}$ is minimized by varying the parameter a of the hydrogenic $1s$ wave function $\psi=(8\pi a)^{-3/2}\exp[-r/(2a)]$, whereas the interaction with the medium, V_{int}, is modeled by a self-energy [8,13,20]. We use for V_{int} the above results, Eq. (1) or (3), for the expected value of self-energy in the presence of charge exchange, assuming that only one (ground) bound state may be formed on the projectile which follows adiabatically the time evolution of the self-energy.

For the sake of computation, we consider the case of He^+ and He^{2+} ions $(Z=2)$ moving in an Al target at the speed v in the range from 2 to 5, where $\Gamma\approx 0.15$, while the equilibrium charge-state distribution may be parameterized by $\phi_1/\phi_2=15.13v^{-3.2}$ [7,12,13]. We have evaluated numerically the instantaneous self-energy in Eq. (1) by means of the so-called plasmon pole approximation (PPA) [22] for dielectric function, whence $\epsilon^{-1}(k,\omega)=1+\omega_p^2/[\omega(\omega+i\gamma)-\omega_k^2]$, where $\omega_k^2=\omega_p^2+3v_F^2k^2/5+k^4/4$, with v_F being Fermi speed, while the damping constant is taken to be $\gamma\to 0^+$. However, the numerical results appear to be very well approximated, at least for sufficiently high projectile speeds v, by Drude model for dielectric function, which follows from PPA by setting $\omega_k=\omega_p$. We therefore use Drude model to evaluate self-energies in Eqs. (1) and (3) in analytical form, which greatly reduces computational effort, especially, for long dwell times T. Choosing $v=3$, we show in Fig. 1(a) the

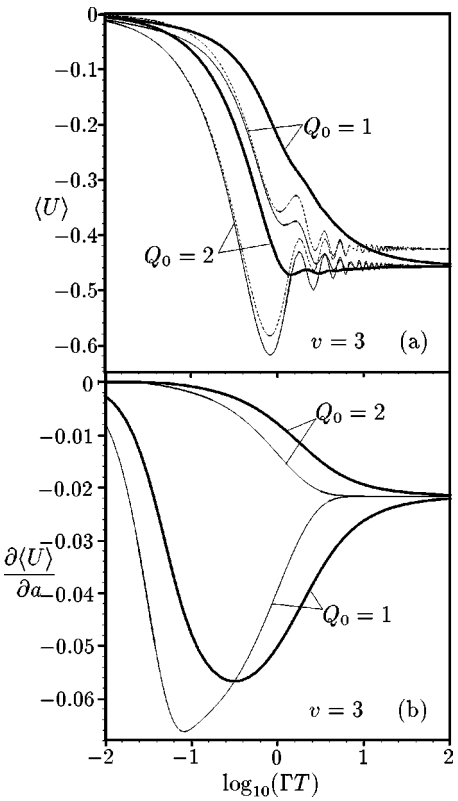

FIG. 1. Expected value of the self-energy (in atomic units) for He ions moving in an Al foil at the speed $v=3$ a.u. versus the logarithm of the reduced dwell time ΓT, with the initial charge states, $Q_0=1$ and 2, and with the unperturbed size of the bound-electron orbit, $a=1/4$ a.u. (a) Thin solid lines, instantaneous self-energy $\langle U(T) \rangle$, Eq. (1); thick solid lines, time-averaged self-energy $\overline{\langle U(T) \rangle}$, Eq. (3); dashed lines, instantaneous self-energy in the point-charge approximation, $a=0$. (b) Derivative of the instantaneous (thin solid lines) and the time-averaged (thick solid lines) self-energy with respect to the bound-electron orbit size parameter a (in atomic units).

dependence on the logarithm of the reduced traversal time ΓT, for both the instantaneous (thin solid line) and the time-averaged (thick solid line) self-energies, evaluated, respectively, from Eqs. (1) and (3) with $a=1/4$, corresponding to the unperturbed $1s$ orbital. In order to demonstrate the effects of the size of the bound-electron orbital, we also show in Fig. 1(a) the instantaneous self-energy (dashed line) evaluated from Eq. (1) with $a=0$, corresponding to the point-charge approximation. The two sets of curves, shown in Fig. 1(a), correspond to two initial charge states, $Q_0=1$ and $Q_0=2$. It is evident from Fig. 1(a) that the self-energy is vanishingly small initially, and that it settles to the equilibrium value only after dwell times of the order of $T \simeq 10\Gamma^{-1}$. Significant dependence on the the initial charge Q_0 is also observed.

As regards the dependence of the self-energy on the variational parameter a, we have found that both Eq. (1) and (3) yield practically linear dependence on a in a broad range from 0 to 1/2, for all ion speeds and dwell times of interest here. It is, therefore, more interesting to examine the derivatives of both the instantaneous and the time-averaged self-energies with respect to a, evaluated at the unperturbed value $a=1/4$, which are displayed in Fig. 1(b) by the thin solid line and the thick solid line, respectively, for ion speed $v=3$. A rather smooth dependence of these derivatives on $\log_{10}(\Gamma T)$ is observed in Fig. 1(b), along with strong effects of differing initial charge states, while the results from Eq. (1) and Eq. (3) appear to be quite similar to each other on a qualitative level. In the subsequent calculations, we use the linearization of Eq. (3) in a about the unperturbed value $a=1/4$, thus, obtaining excellent approximation for the value a which minimizes the total projectile energy $\mathcal{E}_{tot}$ in the range of ion speeds v and dwell times T considered. Moreover, we have found only a minor increase in a, up to about one percent, compared to the unperturbed value $a=1/4$, which is expected for relatively high ion speeds of interest here, and is similar to the results obtained from alternative modeling of the interaction with the target, V_{int} [8,13]. While such a deviation of the variational parameter a from its unperturbed value has practically negligible effects in the subsequent calculation of the ion energy losses, we mention that the variation of this parameter is expected to be stronger for slower ions [7].

III. ENERGY LOSS

In order to derive the expected energy loss of a projectile after the traversal time T, which is assumed to be sufficiently short that the change in the ion velocity $\mathbf{v}$ is negligible, we use the result of the many-body theories for the probability density distribution $\mathcal{P}(E;T)$ of the ion energy loss E after time T [14,15,19],

$$\mathcal{P}(E;T)=\int_{-\infty}^{\infty}\frac{d\tau}{2\pi}\exp[iE\tau-\Phi(\tau;T)], \tag{6}$$

where

$$\Phi(\tau;T)=\int\frac{d^3\mathbf{k}}{(2\pi)^3}\int_0^{\infty}\frac{d\omega}{\pi}(1-e^{-i\omega\tau})\frac{4\pi}{k^2}\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right] \times\left|\int_0^T dt\int d^3\mathbf{r}e^{i(\omega t-\mathbf{k}\cdot\mathbf{r})}\mathcal{D}(\mathbf{r}-\mathbf{v}t,t)\right|^2, \tag{7}$$

with $\epsilon(k,\omega)$ being the dielectric function of the electron gas in the target [22]. Note that the projectile-charge density has been expressed in the target frame of reference in Eq. (7). Upon calculating the mean energy loss of the projectile after the traversal time T as the first moment of the above distribution, $\bar{E}(T)\equiv\int dE\,E\,\mathcal{P}(E;T)=-i\,\partial\Phi/\partial\tau|_{\tau=0}$, we define the expected stopping power after time T by $S_{coh}(T)\equiv\langle\bar{E}(T)\rangle/(vT)$, whence

$$S_{coh}(T)=\frac{2}{v}\int\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{4\pi}{k^2}\int_0^{\infty}d\omega\,\omega \times\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]F(\mathbf{k},\omega-\mathbf{k}\cdot\mathbf{v};T), \tag{8}$$

with the spectral function $F(\mathbf{k},\omega;T)$ of the process $\mathcal{D}(\mathbf{k},t)$ being defined in Eq. (4). The notation $S_{coh}(T)$ suggests that the convolution-type integral over the frequency ω in Eq. (8) gives rise to the coherence effects on the ion energy loss related to the time scale for the target response, implied in the inelastic loss function $\mathrm{Im}[-1/\epsilon(k,\omega)]$, and the time scale for the charge-changing events, contained in the nonstationary time correlation function, Eqs. (2) and (4). More precisely, it is the inherent coupling of the ω- and T-dependence in the spectral function $F(\mathbf{k},\omega;T)$ that affects the mechanisms of the energy deposition in the target, via Eq. (8).

It is worthwhile discussing, at least at the qualitative level, the relation of Eq. (8) with the result of the standard theories of ion stopping, where coherence effects are absent [3,7–9,13]. A critical step in that direction is to consider under what conditions would the ω-integration in Eq. (8) become, at least approximately, decoupled from the T-dependence. In that context, one should notice that the ω-dependence of the spectral function $F(\mathbf{k},\omega;T)$, Eq. (4), is rather broadly structured for, say, $\Gamma T<1$, while it contains only narrow structures with typical widths of the order of Γ and T^{-1} when $\Gamma T>1$, or $\exp(-\Gamma T)\ll 1$, to be more precise. Thus, coherence effects will certainly be pronounced in the pre-equilibrium regime, $\Gamma T<1$, while their persistence for longer dwell times will depend on whether the ω-dependence of the loss function $\mathrm{Im}[-1/\epsilon(k,\omega)]$ may be considered, upon angular integration of the factor $\mathbf{k}\cdot\mathbf{v}$ in Eq. (8), to be broader or narrower than T^{-1} and Γ. Considering collective and/or single-particle excitations of electron gas in metal, one may assert that, for $\Gamma T>1$ and for a sufficiently fast projectile, such that $\Gamma+\omega_p \ll v^2/2$ and $v_F \ll v$, the factor $F(\mathbf{k},\omega-\mathbf{k}\cdot\mathbf{v};T)$ in Eq. (8) would be sufficiently peaked at $\omega=\mathbf{k}\cdot\mathbf{v}$ that one may approximate

$$S_{coh}\approx \frac{1}{v}\int \frac{d^3\mathbf{k}}{(2\pi)^3}\frac{4\pi}{k^2}(\mathbf{k}\cdot\mathbf{v})\mathrm{Im}\left[\frac{-1}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}\right] \times\int_{-\infty}^{\infty} d\omega\, F(\mathbf{k},\omega-\mathbf{k}\cdot\mathbf{v};T). \tag{9}$$

From Eq. (4) we find

$$\int_{-\infty}^{\infty} d\omega\, F(\mathbf{k},\omega;T)=\frac{1}{T}\int_0^T dt\,\langle \mathcal{D}^2(\mathbf{k},t)\rangle, \tag{10}$$

while Eq. (2) gives

$$\langle \mathcal{D}^2(\mathbf{k},t)\rangle=\sum_{j=1}^{2}\rho_j^2(\mathbf{k})\Pi(Q_j,t|Q_0,0), \tag{11}$$

so that the right-hand side of the approximate relation, Eq. (9), is recognized as the standard result [7–9], which we call the incoherent stopping power $S_{inc}(T)$. It may be defined in a manner similar to that of its coherent counterpart, that is, as the time average over the traversal time T, $S_{inc}(T)=T^{-1}\int_0^T dt\langle dE/dx\rangle$, of the expected energy loss per unit path length at time t, $\langle dE/dx\rangle$, given by the incoherent superposition over the instantaneous charge-state distribution, i.e.,

$$\left\langle \frac{dE}{dx}(t)\right\rangle=\sum_{j=1}^{2} S_j\Pi(Q_j,t|Q_0,0), \tag{12}$$

where S_j is the stopping power in the fixed charge state Q_j (with $j=1$ or 2) [22],

$$S_j=\frac{1}{v}\int \frac{d^3\mathbf{k}}{(2\pi)^3}\frac{4\pi}{k^2}|\rho_j(\mathbf{k})|^2(\mathbf{k}\cdot\mathbf{v})\mathrm{Im}\left[\frac{-1}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}\right]. \tag{13}$$

Note that, although Eqs. (9)–(13) do not provide a rigorous procedure of "deriving" the standard result [7–9] from the main result of our work, Eq. (8), they nevertheless demonstrate what physical content is different, and what is related, between the S_{coh} and S_{inc}.

Since data for ion energy losses are often given in terms of the so-called effective ion stopping charge [2,3,8,13,23], we represent our results in a similar manner by defining the coherent and incoherent effective charges, $Q_{coh}=\sqrt{S_{coh}/S_p}$ and $Q_{inc}=\sqrt{S_{inc}/S_p}$, respectively, where the proton stopping power S_p is given by Eq. (13) with $\rho_j(\mathbf{k})=1$. We again consider He^+ and He^{2+} projectiles in Al, with the bound-electron density determined from the procedure outlined in the preceding section, while describing the target response with PPA dielectric function [22]. In Fig. 2, we show both the coherent (thick solid line) and the incoherent (thin solid line) effective charges, Q_{coh} and Q_{inc}, for three ion speeds, $v=2$, 3, and 4, and with the initial charges $Q_0=1$ and $Q_0=2$, versus the logarithm of the reduced traversal time ΓT. Also shown by the dashed lines are the expected ion-charge states at time T, defined by $\langle Q(T)\rangle=Q_1\Pi(Q_1,T|Q_0,0)+Q_2\Pi(Q_2,T|Q_0,0)$, in order to illustrate the time dependence of the weighting factors in Eq. (12), as well as to emphasize the quantitative difference between the effective and the actual ion charges.

In general, one observes rather strong dependence of both Q_{coh} and Q_{inc} on the initial charge Q_0 throughout the whole range of dwell times displayed in Fig. 2. Moreover, one observes that the coherent and incoherent charges are practically the same for long dwell times, such that $\Gamma T>1$, say. This result corroborates our intuitive argument used in the above "derivation," Eqs. (9)–(13), which validates the picture of incoherent ion stopping in the equilibrium regime, for sufficiently fast ions. On the other hand, in the pre-equilibrium regime with the dwell times such that $\Gamma T<1$, marked differences are displayed in Fig. 2 between the coherent and incoherent effective charges. At the earliest stages, the coherent charges are much smaller than the corresponding incoherent charges, and are rather weakly dependent on the ion speed v. A steep increase in Q_{coh} is observed for times up to about $T\simeq\Gamma^{-1}$, giving rise to a nonmonotonic behavior of the coherent charge with the initial charge Q_0

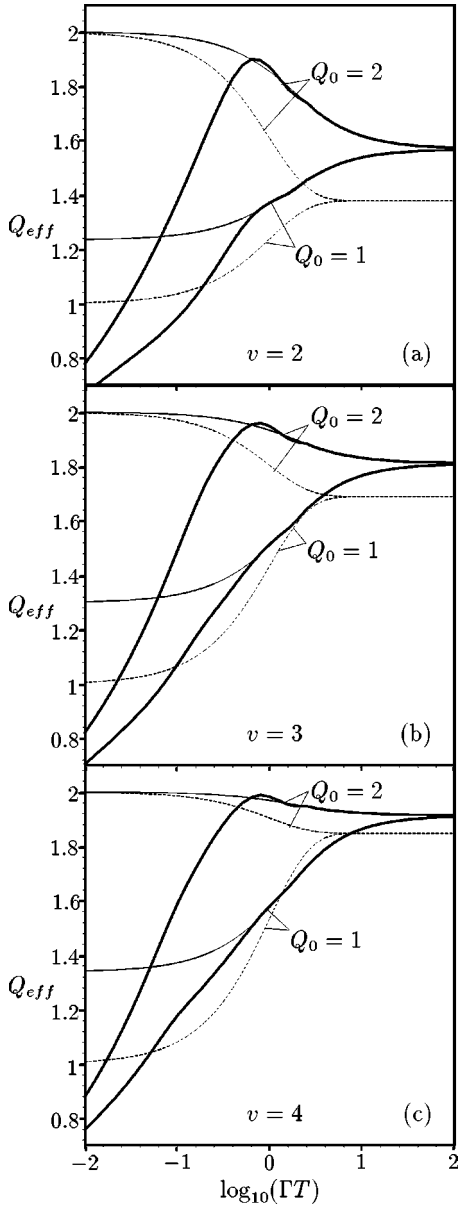

FIG. 2. Effective stopping charges for He ions moving in an Al foil at the speeds (a) $v=2$, (b) $v=3$, and (c) $v=4$ a.u., versus the logarithm of the reduced dwell time ΓT, with the initial charge states, $Q_0=1$ and 2, and with the self-consistent size of the bound-electron orbit. Thick solid lines, coherent effective charge $Q_{coh}=\sqrt{S_{coh}(T)/S_p}$, Eq. (8); thin solid lines, incoherent effective charge $Q_{inc}=\sqrt{S_{inc}(T)/S_p}$ (where S_p is proton stopping power); dashed lines, instantaneous average ion charge $\langle Q(T)\rangle$.

$=2$. We finally note that, for high ion speeds, the charge exchange rates Γ are usually very small [4,5], such that, for the thinnest foils of the experimental interest, one may easily have $\Gamma T \ll 1$, so that the coherence effects, shown in Fig. 2 for the pre-equilibrium dwell times, may become observable.

IV. CONCLUDING REMARKS

In the early stages of penetration through a solid target, the time evolution of the ion-charge state is a random sequence of the charge-changing collisions with the target atoms, which is adequately described as a nonstationary Markov process. Owing to the finite target response time to such sudden and repetitive changes in the projectile-charge density, the instantaneous rate of energy deposition in the target becomes dependent on the history of the charge-state evolution, which is summarized in the temporal autocorrelation function of the Markov process. Such coherent effects in ion energy losses appear to be particularly prominent for short dwell times T through the target, such that $\Gamma T<1$, where Γ is the charge-exchange rate.

By considering hydrogenlike projectiles at the medium-to-high speeds v, we have evaluated the time dependence of the expected value of the projectile self-energy in the presence of charge-exchange processes, and used that result to determine self-consistently the bound-electron density by minimizing the total energy of the projectile. We have further evaluated the expected energy loss after the dwell time T and represented the results for He projectiles in aluminum by means of the effective stopping charge. Significant coherence effects in the energy loss have been found in the pre-equilibrium regime, such that $\Gamma T<1$, when compared to the results obtained from a model where the instantaneous energy-loss rate is given by an incoherent superposition over the stopping powers at fixed charge states, weighted by the instantaneous charge-state distribution. Our results demonstrate that such an incoherent model of ion stopping, which is characteristic of the existing theories of charge-state effects in ion stopping, is clearly adequate when $\Gamma T>1$, at least for fast hydrogenlike ions in metal targets, where the conditions $\Gamma+\omega_p \ll v^2/2$ and $v_F \ll v$ are fulfilled.

It is expected that the coherence effects in the pre-equilibrium ion stopping may be even more pronounced at lower projectile speeds. In order to extend the present theory in that direction, one has to take into account that the number of the relevant projectile charge states may be greater than two, as well as to use more adequate dielectric formalism, including, e.g., the local-field correction. Additional complications in the case of slow ions are related to inaccuracy of the experimental and theoretical data for the relevant electron capture and loss cross sections in the target, as well as to incomplete understanding of the role of surface effects in both the energy loss and the charge-state evolution.

Recent experiments on ion stopping in laser-produced plasmas, or in fully ionized low-density plasmas, have demonstrated the importance of effective ion charges [24], suggesting a need to apply present model to low-density and hot plasma targets with appropriate modifications of dielectric function and cross sections $\sigma_{C,L}$ for such targets. It would be interesting to study effects of decreasing electron density n_e on the coherence effects in ion stopping, knowing that the plasma frequency and the electron transfer rate scale with n_e as $\omega_p \propto n_e^{1/2}$ and $\Gamma \propto n_e$ (neglecting the density effect on cross sections). Finally, it should be mentioned that the coherent effects were obtained in the present work from a description of projectile-charge state evolution as superposition of time-dependent probabilities. A more refined, quantum-mechanical treatment of this problem would employ a super-

position of time-dependent amplitudes, say, by means of a density matrix for projectile-charge-state evolution [25]. Consequently, there remains a question, which should be addressed in future work, as to whether quantum-mechanical coherent effects are less or more pronounced than those studied in the present work.

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[1] H.-D. Betz, Rev. Mod. Phys. **44**, 465 (1972).
[2] G. Schiwietz and P.L. Grande, Nucl. Instrum. Methods Phys. Res. B **175-177**, 125 (2001); P. Sigmund and A. Shinner, *ibid.* **174**, 535 (2001).
[3] N.E.B. Cowern, P.M. Read, C.J. Sofield, L.B. Bridwell, and M.W. Lucas, Phys. Rev. A **30**, 1682 (1984).
[4] H. Ogawa, I. Katayama, H. Ikegami, Y. Haruyama, A. Aoki, M. Tosaki, F. Fukuzawa, K. Yoshida, I. Sugai, and T. Kaneko, Phys. Rev. B **43**, 11370 (1991); H. Ogawa, N. Sakamoto, I. Katayama, Y. Haruyama, M. Saito, K. Yoshida, M. Tosaki, Y. Susuki, and K. Kimura, Nucl. Instrum. Methods Phys. Res. B **132**, 36 (1997).
[5] H. Ogawa, N. Sakamoto, and H. Tsuchida, Nucl. Instrum. Methods Phys. Res. B **164-165**, 279 (2000).
[6] D. Semrad, M. Bergsman, P. Bauer, R. Diez-Muino, and S.A. Arnau, Nucl. Instrum. Methods Phys. Res. B **164-165**, 284 (2000).
[7] F. Sols and F. Flores, Phys. Rev. B **30**, 4878 (1984); A. Arnau, M. Penalba, P.M. Echenique, F. Flores, and R.H. Ritchie, Phys. Rev. Lett. **65**, 1024 (1990).
[8] M. Penalba, A. Arnau, and P. M. Echenique, in *Interaction of Charged Particles with Solids and Surfaces*, Vol. 271 of *NATO Advanced Studies Institute Series B: Physics*, edited by A. Gras-Marti, H. M. Urbassek, N. R. Arista, and F. Flores (Plenum, New York, 1991), p. 541.
[9] P. Sigmund, Nucl. Instrum. Methods Phys. Res. B **69**, 113 (1992).
[10] R. Huber, F. Tauser, A. Brodschelm, M. Bichler, G. Abstreiter, and A. Leitenstorfer, Nature (London) **414**, 286 (2001).
[11] W.-D. Schöne and W. Ekardt, Phys. Rev. B **62**, 13464 (2000).
[12] Y. Haruyama, Y. Kanamori, T. Kido, and F. Fukuzawa, J. Phys. B **15**, 779 (1982).
[13] T. Kaneko, Phys. Rev. A **33**, 1602 (1986).
[14] M. Kato, J. Phys. Soc. Jpn. **55**, 1011 (1986).
[15] A. Bilić, B. Gumhalter, W. Mix, A. Golichowski, S. Tzanev, and K.J. Snowdon, Surf. Sci. **307-309**, 165 (1994).
[16] Z.L. Mišković, S.G. Davison, F.O. Goodman, and W.-K. Liu, Phys. Rev. B **60**, 14478 (1999).
[17] A.K. Prinja, Nucl. Instrum. Methods Phys. Res. B **115**, 187 (1996).
[18] A. Papoulis, *Probability, Random Variables, and Stochastic Processes* (McGraw-Hill, New York, 1991).
[19] Z.L. Mišković, F.O. Goodman, and W.-K. Liu, Nucl. Instrum. Methods Phys. Res. B **193**, 32 (2002).
[20] P.M. Echenique, W. Brandt, and R.H. Ritchie, Phys. Rev. B **33**, 43 (1986).
[21] J. Osma and F.J. Garcia de Abajo, Phys. Rev. A **56**, 2032 (1997).
[22] P.M. Echenique, F. Flores, and R.H. Ritchie, Solid State Phys. **43**, 229 (1990).
[23] W. Brandt and M. Kitagawa, Phys. Rev. B **25**, 5631 (1982).
[24] M. Roth *et al.*, Europhys. Lett. **50**, 28 (2002).
[25] D.G. Arbó, C.O. Reinhold, P. Kürpick, S. Yoshida, and J. Burgdörfer, Phys. Rev. A **60**, 1091 (1999).