Z^3-order theory of quantum inelastic scattering of charges by solids

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Although the nonlinear response of solids in such phenomena as ion slowing and second harmonic generation has been studied since long ago, to our knowledge there has not existed a quantum theory of the inelastic scattering of charges by solids beyond the first Born approximation. In this paper we relate the inelastic cross section in the second Born approximation to the order Z^3 to the quadratic retarded density-response function in the same (but far less trivial) fashion it has been known for the first Born approximation, deriving by this a formula applicable to describe the electron and positron energy-loss spectroscopy. The complete account of recoil is preserved. Our general formalism neither relies on a specific approximation to the dielectric response (such as the random phase approximation) nor is it restricted to scattering by a homogeneous electron gas: it is "exact" in the sense of inclusion of exchange and correlation and is applicable to targets of arbitrary symmetry. Based on this formalism, we perform explicit calculations of the Z^3 contribution to plasmon excitation by electrons and positrons in a simple hydrodynamic model of electron gas and discuss the results, which prove to be instructive in the general case too.

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I. INTRODUCTION

Although the theory of the response of matter to dynamic external perturbations and the theory of scattering of charges by matter are the closely related subjects, they are not identical. Indeed, in the former a target (usually considered a quantum system) is excited by a classical external field, e.g., by the Coulomb field of an ion moving along a well-defined trajectory or by light governed by Maxwell equations. In contrast, in the scattering of electrons or positrons by matter the projectiles themselves are quantum particles and cannot be substituted by an external field. This point becomes particularly clear if one considers an electron moving in a continuous homogenious isotropic medium. If we regard this electron as a classical particle traveling along a definite trajectory, then by virtue of symmetry it cannot deviate from its initial straight path, the only influence of the medium being the continuous slowing of its motion. It is well known, though, that such an electron has a probability to deviate in any direction with the corresponding momentum transfer to the medium, which is entirely the quantum effect.

However, it is well known, too, that the many-body inelastic cross section of electron scattering in the first Born approximation can be expressed in terms of the linear density-response function, the same function which describes the target response to a classical external field. The great practical benefit from such a relationship is that it is the response function to an external field which can be calculated in more or less accurate approximations [e.g., hydrodynamic, random phase (RPA), or time-dependent local density (TDLDA) approximations]; then, the inelastic cross section can be obtained by use of it. To the best of our knowledge this scheme, trivial as it is in the linear case, has been restricted so far to the first Born approximation. As long as a correspondence between the higher-order inelastic cross sections and the nonlinear response functions is found, one can take advantage of the latter, known from the literature in conjunction with other phenomena, to solve the problem of electron and positron energy losses in solids. Examples of systems for which the quadratic response functions are known analytically or can be calculated numerically are the three-[1] and two-[2] dimensional (3D and 2D) electron gas, hydrodynamic,[3] and jellium[4] model surfaces.

The purpose of this paper is to extend the above-mentioned scheme to the second Born approximation. The effects beyond the first Born approximation are important in many ways, the most obvious being maybe that in the linear theory there is no difference between the scattering of charges of opposite sign. We will also show that the quadratic contribution to the plasmon excitation becomes comparable with the linear one near the plasmon excitation threshold. The main result of this paper is a formula for the inelastic differential cross section to order Z^3, where Ze is the charge of an incident distinguishable particle:

$$\frac{d\sigma(\mathbf{p}-\mathbf{k}\leftarrow\mathbf{p})}{d\mathbf{k}}=\frac{-32e^4Z^2\pi^2}{vk^2}\Theta(\omega)\Bigg\{\frac{1}{ek^2}\mathrm{Im}\chi_1(\mathbf{k},\mathbf{k},\omega)+\frac{Z}{2\pi^3}\mathrm{Re}\int\frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\times\int_0^\infty\frac{\Theta(\omega-\omega_1)\nu(\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{q},\omega,\omega_1)+\Theta(\omega_1-\omega)\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\omega)}{\mathbf{vq}-\frac{q^2}{2\mu}-\omega_1+i\eta}d\omega_1\Bigg\}. \quad (1)$$

Here $\mathbf{p}$ is the incident momentum of the charge, μ is its mass, $\mathbf{v}=\mathbf{p}/\mu$ is the incident velocity, $\mathbf{k}$ and $\omega=(p^2-|\mathbf{p}-\mathbf{k}|^2)/2\mu$ are the momentum and the energy loss, respectively, Θ is the Heaviside's step function, and η is the positive infinitesimal. The function ν is defined by the relations

$$\lambda(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)-\chi_2(-\mathbf{k},-\mathbf{q},\mathbf{f},\omega_1-\omega,\omega_1), \quad (2)$$

$$\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\frac{1}{2}[\lambda(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)+\lambda(\mathbf{q},\mathbf{k},\mathbf{f},-\omega,-\omega_1)], \quad (3)$$

and the linear and quadratic density-response functions are defined in the theory of response to an external field by

$$\delta n(\mathbf{q},\omega)=\int\chi_1(\mathbf{q},\mathbf{k},\omega)\phi_{ext}(\mathbf{k},\omega)d\mathbf{k}+\int\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)\phi_{ext}(\mathbf{k},\omega-\omega_1)\times\phi_{ext}(\mathbf{f},\omega_1)d\mathbf{k}d\mathbf{f}d\omega_1, \quad (4)$$

where ϕ_{ext} is the scalar potential of an external electric field perturbing the system, and δn is the induced particle density. All quantities are written in the reciprocal wave-vector-frequency representation. Equations (1)–(3) relate the "exact" many-body cross section to order Z^3 to the "exact" many-body quadratic response function in the sense that they are not restricted to any specific approximation of the latter (i.e., RPA, etc.). In particular, formula(1) is valid (to order Z^3) for scattering near the plasmon excitation threshold, when the momentum of the projectile before and after the scattering differs substantially. Furthermore, no restriction on the nature of the target is being imposed, so Eq. (1) is valid, for example, for spatially confined targets as well as for homogenious 3D or 2D electron gas.

For heavy particles such as ions, which can be considered moving along classical trajectories, when the recoil can be neglected, the problem of relating the cross sections to the response function, we are mainly concerned with here, does not arise, and the detailed nonlinear theory had been developed in Refs. 5 and 6. However, even in this case our approach puts the decay rate and the stopping power into the form when they can be calculated as the total probability and the expectation value of the energy loss, respectively, with the same probability density, which has presented a problem before now.[7]

This paper has the following structure. In Sec. II we write down the inelastic cross section to order Z^3 in terms of the complete set of many-body eigenstates of the target. In Sec. III we reproduce the derivation of the quadratic density-response function to a classical external field. In Sec. IV we derive the relation between the inelastic cross section and density-response functions to order Z^3 [Eq. (1)]. In Sec. V we consider the simplifications arising when the recoil can be neglected. Section VI treats homogeneous isotropic targets. Section VII makes the connection of our theory to the decay rate and stopping power calculations known from the literature. In Sec. VIII we give an example of an explicit calculation in a simple hydrodynamic model of a boundless electron gas. In Sec. IX the results are summarized. The Appendixes contain some useful properties of the response functions and some lengthy derivations.

II. MANY-BODY DIFFERENTIAL CROSS SECTION

Let us consider the inelastic scattering of a charge by a solid. The transition matrix element to second order in the Born series is[8,9]

$$\langle\mathbf{p}',n|T|\mathbf{p},0\rangle=\langle\mathbf{p}',n|V_{cs}+V_{cs}\frac{1}{p^2/2\mu+E_0-\hat{H}_c-\hat{H}_s+i\eta}V_{cs}|\mathbf{p},0\rangle, \quad (5)$$

where $|\mathbf{p},n\rangle$ is the state with projectile of momentum $\mathbf{p}$ and the solid at state $|n\rangle$, E_0 is the ground-state energy of the target, $\hat{H}_c$ and $\hat{H}_s$ are the Hamiltonians of the charge and the solid, respectively, without interaction between them, and V_{cs} is the incident charge-solid interaction,[10]

$$V_{cs}=\sum_j\frac{Ze^2}{|\mathbf{r}-\mathbf{r}_j|},$$

where $\mathbf{r}$ and $\mathbf{r}_j$ are the coordinates of the incident charge and the charges of the solid, respectively and, e is the charge of carriers in the system, negative if electrons. After Fourier transforming

$$V_{cs}=\frac{Ze^2}{2\pi^2}\int \frac{e^{i\mathbf{r}\cdot\mathbf{k}}}{k^2}n_{\mathbf{k}}d\mathbf{k},$$

where

$$n_{\mathbf{k}}=\sum_j e^{-i\mathbf{k}\cdot\mathbf{r}_j}$$

is the particle-density operator, and substituting into Eq. (5), we have

$$\langle \mathbf{p}-\mathbf{k},n|T|\mathbf{p},0\rangle =\frac{Ze^2}{2\pi^2k^2}n_{-\mathbf{k}}^{n0}+\frac{Z^2e^4}{4\pi^4}\int \frac{d\mathbf{q}}{q^2|\mathbf{k}+\mathbf{q}|^2}\times\frac{n_{\mathbf{q}}^{nm}n_{-\mathbf{k}-\mathbf{q}}^{m0}}{[p^2-(\mathbf{p}-\mathbf{k}-\mathbf{q})^2]/2\mu-\omega_{m0}+i\eta},$$

where $\omega_{nm}=E_n-E_m$ and $n_{\mathbf{q}}^{nm}=\langle n|n_{\mathbf{q}}|m\rangle$. Summation over the repeated index m is implied. Without loss of generality, we assume $|n\rangle$ to be real. Then retaining terms to order V_{ext}^3, we have for the differential cross section[8,9]

$$\frac{d\sigma(\mathbf{p}-\mathbf{k}\leftarrow\mathbf{p})}{d\mathbf{k}} =\frac{(2\pi)^4\mu}{p}\sum_n |\langle \mathbf{p}-\mathbf{k},n|T|\mathbf{p},0\rangle|^2\delta(\omega-\omega_{n0}) =\frac{4Z^2e^4\mu}{pk^2}\sum_n\left[\frac{1}{k^2}|n_{\mathbf{k}}^{n0}|^2+\frac{Ze^2}{\pi^2}\sum_m \mathrm{Re}\int\frac{d\mathbf{q}}{q^2|\mathbf{k}+\mathbf{q}|^2}\times\frac{n_{\mathbf{k}}^{0n}n_{\mathbf{q}}^{nm}n_{-\mathbf{k}-\mathbf{q}}^{m0}}{[p^2-(\mathbf{p}-\mathbf{k}-\mathbf{q})^2]/2\mu-\omega_{m0}+i\eta}\right]\delta(\omega-\omega_{n0}). \quad (6)$$

III. MANY-BODY DENSITY-RESPONSE FUNCTION

Let us consider a solid under a time-dependent external potential $\phi_{ext}(\mathbf{r},t)$. Then we can write the equation of motion for the density matrix,[11]

$$i\frac{\partial\rho}{\partial t}=[H_0+e\phi_{ext},\rho], \quad (7)$$

where H_0 is the unperturbed Hamiltonian. Expanding ρ in the external potential,

$$\rho=\rho_0+\rho_1+\rho_2,$$

and substituting into Eq. (7), we have

$$i\frac{\partial\rho_1}{\partial t}=[H_0,\rho_1]+e[\phi_{ext},\rho_0],$$

$$i\frac{\partial\rho_2}{\partial t}=[H_0,\rho_2]+e[\phi_{ext},\rho_1]. \quad (8)$$

Taking the matrix elements of Eqs. (8) between unperturbed states and Fourier transforming with respect to time, we have

$$\langle n|\rho_1(\omega)|m\rangle=e\frac{\delta_{0m}-\delta_{0n}}{\omega-\omega_{nm}+i\eta}\langle n|\phi_{ext}(\omega)|m\rangle \quad \text{(no summation)}, \quad (9)$$

$$\langle n|\rho_2(\omega)|m\rangle=e\frac{\langle n|[\phi_{ext},\rho_1](\omega)|m\rangle}{\omega-\omega_{nm}+i\eta} =e\int\frac{\langle n|[\phi_{ext}(\omega-\omega_1),\rho_1(\omega_1)]|m\rangle}{\omega-\omega_{nm}+i\eta}d\omega_1, \quad (10)$$

where δ_{nm} is Kronecker's symbol. ϕ_{ext} can be Fourier expanded with respect to space coordinates,

$$\phi_{ext}(\omega)=\sum_j \phi_{ext}(\mathbf{r}_j,\omega)=\int \phi_{ext}(\mathbf{q},\omega)n_{-\mathbf{q}}d\mathbf{q}. \quad (11)$$

The particle-density fluctuations are given by

$$\delta n(\mathbf{q},\omega)=\frac{1}{(2\pi)^3}\mathrm{Sp}\{n_{\mathbf{q}}[\rho_1(\omega)+\rho_2(\omega)]\}. \quad (12)$$

Substituting Eq. (9) into Eq. (10), then both of them into Eq. (12), and using Eq. (11) and the definition of the density-response functions (4), we obtain

$$\chi_1(\mathbf{q},\mathbf{k},\omega)=\frac{e}{(2\pi)^3}\sum_n n_{\mathbf{q}}^{0n}n_{-\mathbf{k}}^{n0}\left[\frac{1}{\omega-\omega_{n0}+i\eta}-\frac{1}{\omega+\omega_{n0}+i\eta}\right], \quad (13)$$

$$\tilde{\chi}_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1) =\frac{e^2}{(2\pi)^3}\sum_{n,m}\frac{n_{\mathbf{q}}^{0n}n_{-\mathbf{k}}^{nm}n_{-\mathbf{f}}^{m0}}{(\omega-\omega_{n0}+i\eta)(\omega_1-\omega_{m0}+i\eta)} -\frac{n_{\mathbf{q}}^{mn}n_{-\mathbf{k}}^{n0}n_{-\mathbf{f}}^{0m}}{(\omega-\omega_{nm}+i\eta)(\omega_1-\omega_{0m}+i\eta)} -\frac{n_{\mathbf{q}}^{nm}n_{-\mathbf{k}}^{0n}n_{-\mathbf{f}}^{m0}}{(\omega-\omega_{mn}+i\eta)(\omega_1-\omega_{m0}+i\eta)} +\frac{n_{\mathbf{q}}^{0n}n_{-\mathbf{k}}^{nm}n_{-\mathbf{f}}^{m0}}{(\omega-\omega_{0n}+i\eta)(\omega_1-\omega_{0m}+i\eta)}. \quad (14)$$

Let us note that in Eq. (4) the charge-density fluctuations δn are invariant under the transformation

$$\tilde{\chi}_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)\rightarrow\tilde{\chi}_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)+F(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1),$$

with an arbitrary function F satisfying the relation

$$F(\mathbf{q},\mathbf{f},\mathbf{k},\omega,\omega-\omega_1)=-F(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1).$$

To impose a unique definition of the quadratic response function by Eq. (4), we choose the symmetric form of the latter[12]:

$$\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\frac{1}{2}[\tilde{\chi}_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)+\tilde{\chi}_2(\mathbf{q},\mathbf{f},\mathbf{k},\omega,\omega-\omega_1)]. \tag{15}$$

After substitution of Eq. (14) into Eq. (15), the second term can be combined with the seventh, and the third with the sixth,[13] yielding

$$\begin{aligned}\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)&=\frac{e^2}{2(2\pi)^3}\sum_{n,m}\frac{n_{\mathbf{q}}^{0n}n_{-\mathbf{k}}^{nm}n_{-\mathbf{f}}^{m0}}{(\omega-\omega_{n0}+i\eta)(\omega_1-\omega_{m0}+i\eta)}\\&-\frac{n_{\mathbf{q}}^{mn}n_{-\mathbf{k}}^{n0}n_{-\mathbf{f}}^{0m}}{(\omega_1-\omega_{0m}+i\eta)(\omega-\omega_1-\omega_{n0}+i\eta)}\\&+\frac{n_{\mathbf{q}}^{0n}n_{-\mathbf{k}}^{nm}n_{-\mathbf{f}}^{m0}}{(\omega-\omega_{0n}+i\eta)(\omega_1-\omega_{0m}+i\eta)}\\&+(\mathbf{k}\leftrightarrow\mathbf{f},\omega_1\rightarrow\omega-\omega_1).\end{aligned} \tag{16}$$

Equation (16) is the retarded quadratic density-response function. Its time-ordered counterpart, which we will also need below, is[13]

$$\begin{aligned}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)&=\frac{e^2}{2(2\pi)^3}\sum_{n,m}\frac{n_{\mathbf{q}}^{0n}n_{-\mathbf{k}}^{nm}n_{-\mathbf{f}}^{m0}}{(\omega-\omega_{n0}+i\eta)(\omega_1-\omega_{m0}+i\eta)}\\&-\frac{n_{\mathbf{q}}^{mn}n_{-\mathbf{k}}^{n0}n_{-\mathbf{f}}^{0m}}{(\omega_1-\omega_{0m}-i\eta)(\omega-\omega_1-\omega_{n0}+i\eta)}\\&+\frac{n_{\mathbf{q}}^{0n}n_{-\mathbf{k}}^{nm}n_{-\mathbf{f}}^{m0}}{(\omega-\omega_{0n}-i\eta)(\omega_1-\omega_{0m}-i\eta)}\\&+(\mathbf{k}\leftrightarrow\mathbf{f},\omega_1\rightarrow\omega-\omega_1).\end{aligned} \tag{17}$$

IV. RELATING THE CROSS SECTION TO THE RESPONSE FUNCTION

We want to express the cross section (6) in terms of the response functions (13) and (16). This is straightforward to do with the linear term. By Eq. (13) we have

$$\sum_m |n_{\mathbf{k}}^{m0}|^2\delta(\omega-\omega_{m0})=-\frac{8\pi^2}{e}\Theta(\omega)\mathrm{Im}\chi_1(\mathbf{k},\mathbf{k},\omega). \tag{18}$$

In what follows we are concerned with the quadratic term. Using the definition (3) of the function ν and Eqs. (14) and (15), one can verify directly that[14]

$$\begin{aligned}\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)&=-\frac{e^2}{4\pi}\sum_{m,n}n_{\mathbf{q}}^{m0}n_{-\mathbf{f}}^{n0}n_{-\mathbf{k}}^{nm}[\delta(\omega-\omega_{m0})\delta(\omega_1-\omega_{n0})\\&+\delta(\omega+\omega_{m0})\delta(\omega_1+\omega_{n0})]\\&-n_{-\mathbf{k}}^{m0}n_{-\mathbf{f}}^{n0}n_{\mathbf{q}}^{nm}[\delta(\omega_1-\omega_{n0})\delta(\omega-\omega_{nm})\\&+\delta(\omega_1+\omega_{n0})\delta(\omega+\omega_{nm})].\end{aligned} \tag{19}$$

If $\omega_1>0$, then Eq. (19) yields

$$\begin{aligned}\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1>0)&=-\frac{e^2}{4\pi}\sum_{m,n}[n_{\mathbf{q}}^{m0}n_{-\mathbf{f}}^{n0}n_{-\mathbf{k}}^{nm}\delta(\omega-\omega_{m0})\\&-n_{-\mathbf{k}}^{m0}n_{-\mathbf{f}}^{n0}n_{\mathbf{q}}^{nm}\delta(\omega-\omega_{nm})]\delta(\omega_1-\omega_{n0})\end{aligned} \tag{20}$$

or, using the properties of Dirac's δ function,

$$\begin{aligned}\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1>0)&=-\frac{e^2}{4\pi}\sum_{m,n}[n_{\mathbf{q}}^{m0}n_{-\mathbf{f}}^{n0}n_{-\mathbf{k}}^{nm}\delta(\omega-\omega_{m0})\\&-n_{-\mathbf{k}}^{m0}n_{-\mathbf{f}}^{n0}n_{\mathbf{q}}^{nm}\delta(\omega-\omega_1+\omega_{m0})]\delta(\omega_1-\omega_{n0}).\end{aligned} \tag{21}$$

This gives

$$\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega>\omega_1,\omega_1>0)=-\frac{e^2}{4\pi}\sum_{m,n}n_{\mathbf{q}}^{m0}n_{-\mathbf{f}}^{n0}n_{-\mathbf{k}}^{nm}\delta(\omega-\omega_{m0})\delta(\omega_1-\omega_{n0}). \tag{22}$$

Performing permutations $\mathbf{q}\leftrightarrow-\mathbf{f},\omega\leftrightarrow\omega_1$, we have from Eq. (20)

$$\begin{aligned}\nu(-\mathbf{f},\mathbf{k},-\mathbf{q},\omega_1,\omega>0)&=-\frac{e^2}{4\pi}\sum_{m,n}[n_{-\mathbf{f}}^{m0}n_{\mathbf{q}}^{n0}n_{-\mathbf{k}}^{nm}\delta(\omega_1-\omega_{m0})\\&-n_{-\mathbf{k}}^{m0}n_{\mathbf{q}}^{n0}n_{-\mathbf{f}}^{nm}\delta(\omega_1-\omega+\omega_{m0})]\delta(\omega-\omega_{n0})\end{aligned} \tag{23}$$

and then

$$\nu(-\mathbf{f},\mathbf{k},-\mathbf{q},\omega_1>\omega,\omega>0)=-\frac{e^2}{4\pi}\sum_{m,n}n_{-\mathbf{f}}^{m0}n_{\mathbf{q}}^{n0}n_{-\mathbf{k}}^{nm}\delta(\omega_1-\omega_{m0})\delta(\omega-\omega_{n0}). \tag{24}$$

Interchanging the dummy summation indices m and n in Eq. (24) and combining it with Eq. (22) we have

$$\begin{aligned}\sum_{m,n}n_{\mathbf{q}}^{m0}n_{-\mathbf{f}}^{n0}n_{-\mathbf{k}}^{nm}\delta(\omega-\omega_{m0})\delta(\omega_1-\omega_{n0})&=-\frac{4\pi}{e^2}\Theta(\omega)\Theta(\omega_1)[\Theta(\omega-\omega_1)\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)\\&+\Theta(\omega_1-\omega)\nu(-\mathbf{f},\mathbf{k},-\mathbf{q},\omega_1,\omega)].\end{aligned} \tag{25}$$

Now we can also write

$$\sum_{m,n} \frac{n_{\mathbf{q}}^{m0} n_{-\mathbf{f}}^{n0} n_{-\mathbf{k}}^{nm}}{\omega-\omega_{m0}+i\eta}\delta(\omega_1-\omega_{n0}) = \int_{-\infty}^{\infty}\sum_{m,n}\frac{n_{\mathbf{q}}^{m0} n_{-\mathbf{f}}^{n0} n_{-\mathbf{k}}^{nm}}{\omega-\omega_2+i\eta}\delta(\omega_2-\omega_{m0})\delta(\omega_1-\omega_{n0})d\omega_2$$

$$= -\frac{4\pi}{e^2}\Theta(\omega_1)\int_0^{\infty}\frac{\Theta(\omega_1-\omega_2)\nu(-\mathbf{f},\mathbf{k},-\mathbf{q},\omega_1,\omega_2)+\Theta(\omega_2-\omega_1)\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega_2,\omega_1)}{\omega-\omega_2+i\eta}d\omega_2. \tag{26}$$

Finally, using Eqs. (6), (18), and (26), and performing the integration variable substitution $\mathbf{q}\to\mathbf{q}-\mathbf{k}$, we see that the main equation (1) is proved.

V. NEGLECT OF RECOIL

For high incident velocities or heavy projectiles the recoil can be neglected, in which case formula (1)can be simplified. We will restrict our consideration to systems with an inversion center. Then $i\eta$ in Eq. (1) can be dropped, since ν is real, and the principal value of the integral can be implied instead. In this case we can neglect $q^2/2\mu$ in the denominator, and put $\omega=\mathbf{vk}$. Then the integrals in the Z^3 term of Eq. (1) can be written as

$$\int\frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\left[\int_0^{\mathbf{vk}}\frac{\nu(\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{q},\mathbf{vk},\omega_1)}{\mathbf{vq}-\omega_1}d\omega_1+\int_{\mathbf{vk}}^{\infty}\frac{\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\mathbf{vk})}{\mathbf{vq}-\omega_1}d\omega_1\right]. \tag{27}$$

Using the property (A2), then making the integration variables substitutions $\mathbf{q}\to\mathbf{k}-\mathbf{q}$, $\omega_1\to\mathbf{vk}-\omega_1$, we can write for the second integral

$$\int\frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\int_{\mathbf{vk}}^{\infty}\frac{\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\mathbf{vk})}{\mathbf{vq}-\omega_1}d\omega_1=\int\frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\int_{-\infty}^{0}\frac{\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\mathbf{vk})}{\mathbf{vq}-\omega_1}d\omega_1,$$

and then expression (27) can be written as

$$\int\frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\left[\int_0^{\mathbf{vk}}\frac{\nu(\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{q},\mathbf{vk},\omega_1)-\frac{1}{2}\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\mathbf{vk})}{\mathbf{vq}-\omega_1}d\omega_1+\frac{1}{2}\int_{-\infty}^{\infty}\frac{\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\mathbf{vk})}{\mathbf{vq}-\omega_1}d\omega_1\right]. \tag{28}$$

We can also write

$$\int\frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\int_0^{\mathbf{vk}}\frac{\nu(\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{q},\mathbf{vk},\omega_1)-\frac{1}{2}\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\mathbf{vk})}{\mathbf{vq}-\omega_1}d\omega_1 \tag{29}$$

$$=\int\frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\int_0^{\mathbf{vk}}\frac{\nu(\mathbf{k},\mathbf{q},\mathbf{k}-\mathbf{q},\mathbf{vk},\mathbf{vk}-\omega_1)+\frac{1}{2}\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\mathbf{vk})}{\mathbf{vq}-\omega_1}d\omega_1 \tag{30}$$

$$=\int\frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\int_0^{\mathbf{vk}}\frac{-\nu(\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{q},\mathbf{vk},\omega_1)+\frac{1}{2}\nu(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\omega_1,\mathbf{vk})}{\mathbf{vq}-\omega_1}d\omega_1, \tag{31}$$

where we have first used the property (A1), then have again performed the substitution $\mathbf{q}\to\mathbf{k}-\mathbf{q}$, $\omega_1\to\mathbf{vk}-\omega_1$ in the first term of Eq. (30). Comparing Eqs. (29) and (31) we see that the first integral in the square brackets of Eq. (28) is zero. Now using Eq. (B2) of Appendix B, we can write Eq. (27) as

$$\pi \mathrm{Im} \int \frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2} [\chi_2(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\mathbf{vq},\mathbf{vk}) + \chi_2(\mathbf{q}-\mathbf{k},\mathbf{q},-\mathbf{k},\mathbf{v}(\mathbf{k}-\mathbf{q}),\mathbf{vk})]. \tag{32}$$

Making again the substitution $\mathbf{q}\to\mathbf{k}-\mathbf{q}$ in the second term of this expression, we see that the two terms are equal, and we finally obtain, neglecting recoil,

$$\frac{d\sigma(\mathbf{p}-\mathbf{k}\leftarrow\mathbf{p})}{d\mathbf{k}} = \frac{-32e^4Z^2\pi^2}{vk^2}\Theta(\mathbf{vk})\left[\frac{1}{ek^2}\mathrm{Im}\chi_1(\mathbf{k},\mathbf{k},\mathbf{vk}) + \frac{Z}{\pi^2}\int \frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(-\mathbf{q},\mathbf{k}-\mathbf{q},-\mathbf{k},\mathbf{vq},\mathbf{vk})\right]. \tag{33}$$

VI. CASE OF HOMOGENEOUS MEDIUM

If the target is spatially homogeneous, then

$$\delta n(\mathbf{q},\omega) = \chi_1(\mathbf{q},\omega)\phi_{ext}(\mathbf{q},\omega) + \int \chi_2(\mathbf{q},\mathbf{f},\omega,\omega_1)\phi_{ext}(\mathbf{q}-\mathbf{f},\omega-\omega_1) \times \phi_{ext}(\mathbf{f},\omega_1)d\mathbf{f}d\omega_1. \tag{34}$$

The corresponding reduced response functions are collected in Eqs. (A4)–(A11) of Appendix A. Using the definition of the function ν, it is easy to show that the latter is the real function for systems with an inversion center. Then Eq. (1) together with Eqs. (A4) and (A10) gives for the differential cross section

$$\frac{d\sigma(\mathbf{p}-\mathbf{k}\leftarrow\mathbf{p})}{d\mathbf{k}} = \frac{-4\Omega e^4Z^2}{\pi vk^2}\Theta(\omega)\left\{\frac{1}{ek^2}\mathrm{Im}\chi_1(\mathbf{k},\omega) + \frac{Z}{2\pi^3}\int \frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2} \times \int_0^\infty \frac{\Theta(\omega-\omega_1)\nu(\mathbf{k},\mathbf{q},\omega,\omega_1)+\Theta(\omega_1-\omega)\nu(-\mathbf{q},-\mathbf{k},\omega_1,\omega)}{\mathbf{vq}-\frac{q^2}{2\mu}-\omega_1}d\omega_1\right\}, \tag{35}$$

where the principal value of the integral is implied. We have substituted the volume of the target $\Omega/(2\pi)^3$ for the $\delta(\mathbf{q}=0)$. Without the coefficient Ω this quantity gives the inverse mean free path with respect to the given momentum transfer. Similarly to Eq. (33), in the homogeneous case we have an approximation for the case of neglect of the recoil:

$$\frac{d\sigma(\mathbf{p}-\mathbf{k}\leftarrow\mathbf{p})}{d\mathbf{k}} = \frac{-4\Omega e^4Z^2}{\pi vk^2}\Theta(\mathbf{vk})\left[\frac{1}{ek^2}\mathrm{Im}\chi_1(\mathbf{k},\mathbf{vk}) + \frac{Z}{\pi^2}\int \frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk})\right]. \tag{36}$$

VII. DECAY RATE AND STOPPING POWER

Let us make a connection to the decay rate and stopping power calculations known from the literature for an homogenious electron gas. We can easily write these quantities in terms of the cross section with neglect of the recoil:

$$\tau^{-1} = \frac{v}{\Omega}\int \frac{d\sigma(\mathbf{p}-\mathbf{k}\leftarrow\mathbf{p})}{d\mathbf{k}}d\mathbf{k}, \tag{37}$$

$$-\frac{dE_{\mathrm{kin}}}{dx} = \frac{1}{\Omega}\int \mathbf{vk}\frac{d\sigma(\mathbf{p}-\mathbf{k}\leftarrow\mathbf{p})}{d\mathbf{k}}d\mathbf{k}. \tag{38}$$

Substituting Eq. (36) into Eqs. (37) and (38), we have

$$\tau^{-1} = \frac{-4e^4Z^2}{\pi}\int d\mathbf{k}\frac{\Theta(\mathbf{vk})}{k^2}\left[\frac{1}{ek^2}\mathrm{Im}\chi_1(\mathbf{k},\mathbf{vk}) + \frac{Z}{\pi^2}\int \frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk})\right], \tag{39}$$

$$-\frac{dE_{\mathrm{kin}}}{dx} = \frac{-4e^4Z^2}{\pi v}\int d\mathbf{k}\frac{\mathbf{vk}\Theta(\mathbf{vk})}{k^2}\left[\frac{1}{ek^2}\mathrm{Im}\chi_1(\mathbf{k},\mathbf{vk}) + \frac{Z}{\pi^2}\int \frac{d\mathbf{q}}{q^2|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk})\right]. \tag{40}$$

The purpose of this section is to demonstrate that Eqs. (39) and (40) are equivalent to Eqs. (6) and (15) of Ref. 7, respectively, which in our notation are[15]

$$\tau^{-1}=\frac{-4e^4Z^2}{\pi}\int d\mathbf{q}\frac{\Theta(\mathbf{vq})}{k^2}\left[\frac{1}{eq^2}\mathrm{Im}\chi_1(\mathbf{q},\mathbf{vq})+\frac{Z}{3\pi^2}\int\frac{d\mathbf{k}}{k^2|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk})\right], \quad (41)$$

$$-\frac{dE_{\mathrm{kin}}}{dx}=\frac{-4e^4Z^2}{\pi v}\int d\mathbf{q}\frac{\mathbf{vq}\Theta(\mathbf{vq})}{q^2}\left[\frac{1}{eq^2}\mathrm{Im}\chi_1(\mathbf{q},\mathbf{vq})+\frac{Z}{2\pi^2}\int\frac{d\mathbf{k}}{k^2|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk})\right], \quad (42)$$

where χ_2^{TO} is the time-ordered response function (17). The proof is given in Appendix C.

Equations (37) and (38) [or Eqs. (39) and (40)] are from the very beginning put into the form when the stopping power can be obtained from the decay rate by introducing the energy transfer into the integrand. This is not the case with the same quantities in the form of Eqs. (41) and (42), which fact has presented a problem before now.[7]

VIII. EXAMPLE AND DISCUSSION

As the simplest possible but instructive example, let us consider the hydrodynamic model of homogeneous electron gas in the spirit of Ref. 16 but without the pressure term. For the linear response, this model yields a Drude dielectric function without the spatial dispersion. The equations governing the motion are

$$\frac{d\mathbf{v}}{dt}=\frac{\partial\mathbf{v}}{\partial t}+(\mathbf{v}\nabla)\mathbf{v}=-\frac{e}{m}\nabla\phi, \quad (43)$$

$$\frac{\partial n}{\partial t}+\nabla(n\mathbf{v})=0, \quad (44)$$

$$\Delta(\phi-\phi_{ext})=-4\pi en, \quad (45)$$

which are Newton's law, the continuity equation, and Poisson's equation, respectively. Here $\mathbf{v}$, n, ϕ, and ϕ_{ext} are the velocity and density of the electron gas, the total, and the external scalar potentials, respectively. Expanding equations (43)–(45) in powers of ϕ_{ext}, we have to first order

$$\frac{\partial\mathbf{v}_1}{\partial t}=-\frac{e}{m}\nabla\phi_1,$$

$$\frac{\partial n_1}{\partial t}+n_0\nabla\mathbf{v}_1=0,$$

$$\Delta(\phi_1-\phi_{ext})=-4\pi en_1, \quad (46)$$

and to second order

$$\frac{\partial\mathbf{v}_2}{\partial t}+(\nabla\mathbf{v}_1)\mathbf{v}_1=-\frac{e}{m}\nabla\phi_2,$$

$$\frac{\partial n_2}{\partial t}+n_0\nabla\mathbf{v}_2+\nabla(n_1\mathbf{v}_1)=0,$$

$$\Delta\phi_2=-4\pi en_2. \quad (47)$$

After the Fourier transform, we find from Eqs. (46)

$$n_1(\mathbf{q},\omega)=\frac{q^2}{4\pi e}\left[\frac{1}{\epsilon(\omega)}-1\right]\phi_{ext}(\mathbf{q},\omega),$$

$$\mathbf{v}_1(\mathbf{q},\omega)=\frac{e\mathbf{q}}{\omega m\epsilon(\omega)}\phi_{ext}(\mathbf{q},\omega), \quad (48)$$

and hence

$$\chi_1(q,\omega)=\frac{q^2}{4\pi e}\left[\frac{1}{\epsilon(\omega)}-1\right],$$

$$\mathrm{Im}\chi_1(q,\omega)=-\frac{q^2\omega_p}{8e}\delta(\omega-\omega_p),$$

where $\epsilon(\omega)$ is the Drude dielectric function:

$$\epsilon(\omega)=1-\frac{\omega_p^2}{(\omega+i\eta)^2},\omega_p=\sqrt{\frac{4\pi e^2n_0}{m}}.$$

Substituting the solutions of the linear problem (48) into Eqs. (47), we obtain for the quadratic response

$$\tilde{\chi}_2(\mathbf{q},\mathbf{f},\omega,\omega_1)=\frac{\omega_p^2(\omega-\omega_1)[(\mathbf{fq})^2\omega_1+f^2q^2\omega-f^2\mathbf{fq}(\omega+\omega_1)]}{4\pi m[(\omega+i\eta)^2-\omega_p^2][(\omega-\omega_1+i\eta)^2-\omega_p^2][(\omega_1+i\eta)^2-\omega_p^2]},$$

and then, by use of the definitions (A7), (A9), and (A11), it is straightforward to show that

$$\nu(\mathbf{q},\mathbf{f},\omega,\omega_1)=\frac{\pi}{48\ m}\,\delta(\omega_1-\omega_p)[3(\mathbf{fq}-f^2)q^2\delta(\omega)\times[-4(\mathbf{fq})^2+3\mathbf{fq}q^2+f^2q^2]\delta(\omega-2\omega_p)+3\mathbf{fq}(-2\mathbf{fq}+f^2+q^2)\delta(\omega-\omega_p)].$$

For the cross section we then have by virtue of Eq. (35)

$$\frac{d\sigma(\mathbf{p}-\mathbf{k}\leftarrow\mathbf{p})}{d\mathbf{k}}=\frac{Z^2e^2\Omega}{2\pi v k^2}\left\{\omega_p\delta(\omega-\omega_p)+\frac{Ze^2}{12\pi^2 m}\{[A(2\omega_p)+B(\omega_p)]\times\delta(\omega-\omega_p)+C(\omega_p)\delta(\omega-2\omega_p)\}\right\}, \tag{49}$$

where

$$\begin{pmatrix}A\\B\\C\end{pmatrix}=\int\frac{d\mathbf{q}}{q^2|\mathbf{q}-\mathbf{k}|^2\left(\mathbf{vq}-\frac{q^2}{2\mu}-\omega\right)}\times\begin{pmatrix}q^2k^2+3\mathbf{qk}q^2-4(\mathbf{qk})^2\\-3\mathbf{qk}|\mathbf{q}-\mathbf{k}|^2\\q^2k^2+3(\mathbf{qk})k^2-4(\mathbf{qk})^2\end{pmatrix}. \tag{50}$$

Equation (49) contains the single-plasmon excitation in both Z^2 and Z^3 orders, as well as the double-plasmon excitation. Because of the hydrodynamic model, it does not contain the single-particle excitations.

Let us note that the double-plasmon excitation contribution to order Z^2 has been recently reported.[17,18] This effect, which is the consequence of the singularity at $\omega=2\omega_p$ in the high-frequency tail of the imaginary part of the linear dielectric function,[19] is due to the exchange and correlation and is, of cause, beyond our simple hydrodynamic example. Without this term, the Z expansion of the double-plasmon excitation probability starts from the Z^3 term. It is evident that if this term is positive for electrons then it is negative for positrons and vice versa. This situation just demonstrates the logical breakdown of the hydrodynamical model at $\omega=2\omega_p$. The more elaborate RPA approximation is not likely to pass this test for consistency either, since it yields no Z^2 contribution to the double-plasmon excitation probability too.

On the contrary, the Z^3 contribution for the single plasmon is not contradictory and it can give substantial corrections to the Z^2 term. The same can be supposed to be true for single particle-hole excitations, when the latter are allowed by the model of the response function, which evidently is not the case in this simple example. In Figs. 1 and 2 we plot the plasmon excitation intensities versus the incident particle energy obtained by Eqs. (49) and (50) for electron ($Z=1$) and

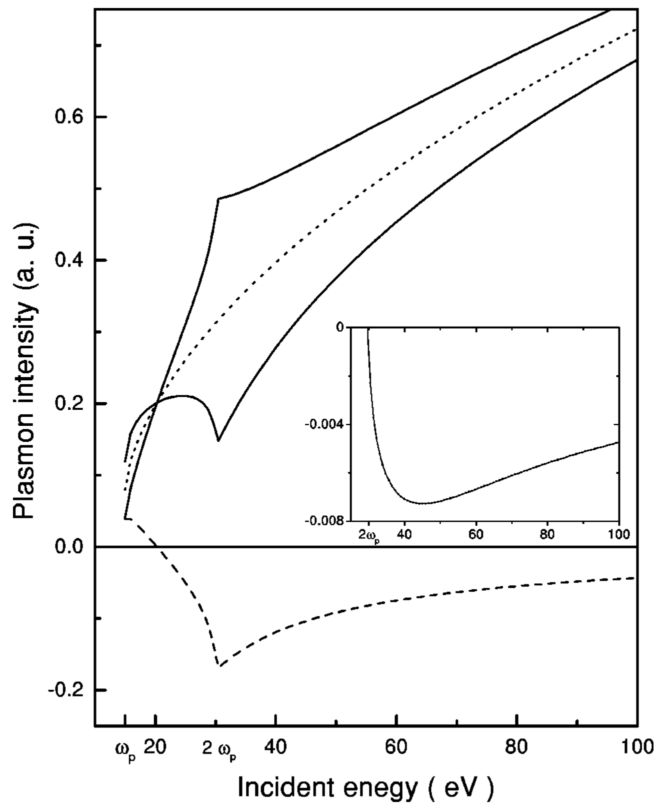

FIG. 1. The plasmon excitation intensity vs the incident energy for the forward scattering. A plasmon energy of 15 eV is chosen. The dotted and dashed lines are the Z^2 and Z^3 contributions, respectively. Two solid lines mainly below (above) the Z^2 contribution are the sum (difference) of the Z^2 and Z^3 terms, corresponding to electron (positron) scattering. The inset shows the double-plasmon intensity, which is the measure of the (in)accuracy of the simple hydrodynamic model of this example (see the text).

positron ($Z=-1$) scattering. Appendix D contains an analytical evaluation of integrals (50) in the case of scattering forward. The inset in Fig. 1 shows the strength of the Z^3 contribution to the double plasmon excitation in this model. Although it happens to be negative for electrons in the chosen range of incident velocities, the comparatively small magnitude of this term shows that the simple hydrodynamic model of this example is not strongly contradictory in this respect.

IX. CONCLUSIONS

In the framework of many-body scattering theory, we have expressed the inelastic differential cross section of a charge scattered by an ensemble of charges to Z^3 order in the second Born approximation in terms of the quadratic retarded density-response function of the target. The latter response function is known from the literature for a number of systems of physical significance, such as 2D and 3D electron gas and jellium model surfaces. The formalism we have developed allows direct application of the knowledge of these response functions to produce the inelastic cross sections of charges interacting with solids. We have supplemented the formal theory with an explicit example of scattering by elec-

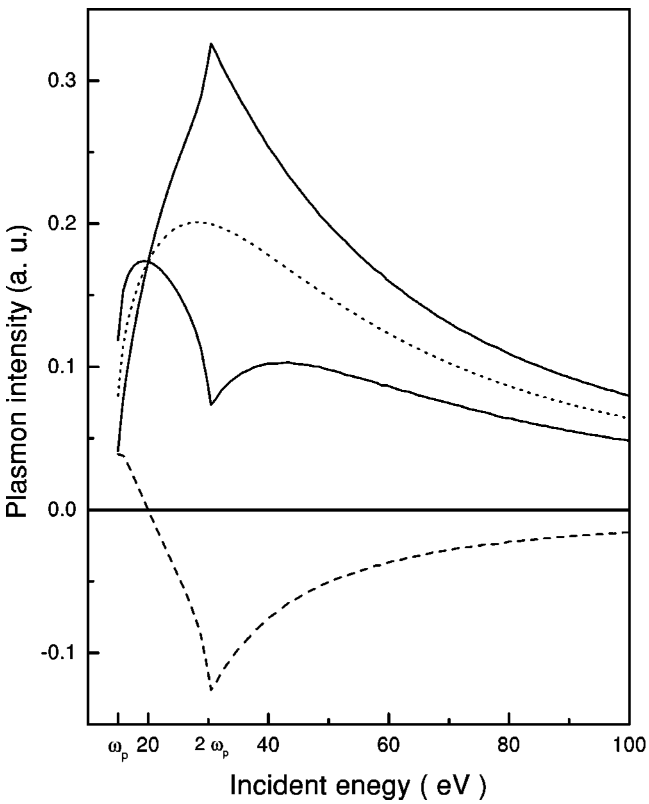

FIG. 2. The same as Fig. 1 but with an angle of scattering of 15°.

tron gas within hydrodynamic model, which demonstrates the importance of nonlinear corrections near the plasmon excitation threshold. The calculations of electron and positron scattering by 3D and 2D electron gas by use of the above theory in the RPA is now in progress.

Note added in proof. In a recent paper[20] various approaches to the decay rate and the stopping power of the homogeneous 3D electron gas have been developed. Apart from our treating the targets not necessarily homogeneous, we see the main difference with Ref. 20 in that our differential cross sections (or transition probabilities) are "exact" to order Z^3, while Ref. 20, although includes exchange and correlation, still uses approximate ones. Also we would like to point out in conjunction with discussion in Ref. 20 that our Eqs. (39) and (40) provide the explicit form to express the decay rate and stopping power, respectively, to order Z^3 in terms of the retarded response functions, where the latter can be obtained from the former by insertion of the energy transfer inside the integrand.

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APPENDIX A: SOME PROPERTIES OF THE RESPONSE FUNCTIONS

The following properties of the ν function are the direct consequences of the definitions (2), (3), and (15):

$$\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\nu(-\mathbf{f},\mathbf{k},-\mathbf{q},\omega_1,\omega)+\nu(\mathbf{q},\mathbf{f},\mathbf{k},\omega,\omega-\omega_1), \tag{A1}$$

$$\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=-\nu(-\mathbf{k},-\mathbf{q},\mathbf{f},\omega_1-\omega,\omega_1), \tag{A2}$$

$$\nu(\mathbf{q},\mathbf{k},\mathbf{f},-\omega,-\omega_1)=\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1). \tag{A3}$$

Using the definitions (15), (2), and (3) we can write in the case of an homogeneous medium

$$\chi_1(\mathbf{q},\mathbf{k},\omega)=\delta(\mathbf{q}-\mathbf{k})\chi_1(\mathbf{q},\omega), \tag{A4}$$

$$\tilde{\chi}_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\delta(\mathbf{k}+\mathbf{f}-\mathbf{q})\tilde{\chi}_2(\mathbf{q},\mathbf{f},\omega,\omega_1), \tag{A5}$$

$$\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\delta(\mathbf{k}+\mathbf{f}-\mathbf{q})\chi_2(\mathbf{q},\mathbf{f},\omega,\omega_1), \tag{A6}$$

$$\chi_2(\mathbf{q},\mathbf{f},\omega,\omega_1)=\frac{1}{2}[\tilde{\chi}_2(\mathbf{q},\mathbf{f},\omega,\omega_1)+\tilde{\chi}_2(\mathbf{q},\mathbf{q}-\mathbf{f},\omega,\omega-\omega_1)], \tag{A7}$$

$$\lambda(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\delta(\mathbf{k}+\mathbf{f}-\mathbf{q})\lambda(\mathbf{q},\mathbf{f},\omega,\omega_1), \tag{A8}$$

$$\lambda(\mathbf{q},\mathbf{f},\omega,\omega_1)=\chi_2(\mathbf{q},\mathbf{f},\omega,\omega_1)-\chi_2(\mathbf{f}-\mathbf{q},\mathbf{f},\omega_1-\omega,\omega_1), \tag{A9}$$

$$\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\delta(\mathbf{k}+\mathbf{f}-\mathbf{q})\nu(\mathbf{q},\mathbf{f},\omega,\omega_1), \tag{A10}$$

$$\nu(\mathbf{q},\mathbf{f},\omega,\omega_1)=\frac{1}{2}[\lambda(\mathbf{q},\mathbf{f},\omega,\omega_1)+\lambda(\mathbf{q},\mathbf{f},-\omega,-\omega_1)]. \tag{A11}$$

The properties

$$\chi_2(\mathbf{q},\mathbf{q}-\mathbf{f},\omega,\omega-\omega_1)=\chi_2(\mathbf{q},\mathbf{f},\omega,\omega_1), \tag{A12}$$

$$\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{q}-\mathbf{f},\omega,\omega-\omega_1)=\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{f},\omega,\omega_1) \tag{A13}$$

are the direct consequence of the symmetrization (15). Using Eqs. (16) and (17) it is straightforward to show that for systems with an inversion center

$$\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\chi_2(-\mathbf{q},-\mathbf{k},-\mathbf{f},\omega,\omega_1), \tag{A14}$$

$$\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\chi_2^{\mathrm{TO}}(-\mathbf{q},-\mathbf{k},-\mathbf{f},\omega,\omega_1), \tag{A15}$$

and the following properties are valid in the general case:

$$\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=-\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},-\omega,-\omega_1), \tag{A16}$$

$$\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=+\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{f},-\omega,-\omega_1). \tag{A17}$$

The following properties hold in the general and homogenious cases, respectively, for χ_2^{TO} *but not for* χ_2:

$$\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\chi_2^{\mathrm{TO}}(-\mathbf{k},-\mathbf{q},\mathbf{f},\omega_1-\omega,\omega_1), \tag{A18}$$

$$\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{f},\omega,\omega_1)=\chi_2^{\mathrm{TO}}(\mathbf{f}-\mathbf{q},\mathbf{f},\omega_1-\omega,\omega_1). \tag{A19}$$

APPENDIX B: DERIVATION OF FORMULA (32)

Let us write down the explicit form of ν according to definitions (2) and (3):

$$\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)=\frac{1}{2}[\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)+\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},-\omega,-\omega_1) -\chi_2(-\mathbf{k},-\mathbf{q},\mathbf{f},\omega-\omega_1,-\omega_1) -\chi_2(-\mathbf{k},-\mathbf{q},\mathbf{f},\omega_1-\omega,\omega_1)]. \quad \text{(B1)}$$

It can be seen from Eqs. (14) and (15) that in Eq. (B1) the first and third terms are analytical functions in ω in the upper complex half-plane, while the other two terms are analytical in the lower complex half-plane. Closing the integration contour in appropriate half-planes and using the property (A16), we then have

$$\int_{-\infty}^{\infty}\frac{\nu(\mathbf{q},\mathbf{k},\mathbf{f},\omega,\omega_1)}{\omega_2-\omega}d\omega=2\pi\mathrm{Im}[\chi_2(\mathbf{q},\mathbf{k},\mathbf{f},\omega_2,\omega_1) +\chi_2(-\mathbf{k},-\mathbf{q},\mathbf{f},\omega_1-\omega_2,\omega_1)]. \quad \text{(B2)}$$

APPENDIX C: PROOF OF EQUIVALENCE OF Eqs. (39), (40) AND (41), (42), RESPECTIVELY

To establish the equivalence of Eqs. (40) and (42), we first note that the Θ function can be dropped from either with simultaneous introduction of the factor 1/2 because of the integral over $\mathbf{q}$ ($\mathbf{k}$) being an even function of $\mathbf{k}$ ($\mathbf{q}$), respectively. Then we can write

$$\int d\mathbf{k}d\mathbf{q}\frac{\mathbf{vk}}{k^2q^2|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}) =\frac{1}{2}\int d\mathbf{k}d\mathbf{q}\frac{\mathbf{vq}}{k^2q^2|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}),$$

the last equation being readily verified by the variable substitution $\mathbf{k}\to\mathbf{q}-\mathbf{k}$ on the left-hand side. This concludes the proof of equivalence of the Eqs. (40) and (42).

Comparing Eqs. (16) and (17) we see that $\chi_2=\chi_2^{TO}$ for $\omega>\omega_1>0$. Then we can write

$$\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})\Theta[\mathbf{v}(\mathbf{q}-\mathbf{k})]}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}) =\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})\Theta[\mathbf{v}(\mathbf{q}-\mathbf{k})]}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}). \quad \text{(C1)}$$

In the left-hand side of Eq. (C1) we write

$$\Theta(\mathbf{vk})\Theta[\mathbf{v}(\mathbf{q}-\mathbf{k})]=[1-\Theta(-\mathbf{vk})]\{1-\Theta[-\mathbf{v}(\mathbf{q}-\mathbf{k})]\}.$$

Expanding the right-hand side of the latter equation and taking advantage of Eqs. (A12) and (A16), we have

$$\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})\Theta[\mathbf{v}(\mathbf{q}-\mathbf{k})]}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}) =\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}). \quad \text{(C2)}$$

For the right-hand side of Eq. (C1) we can write the sequence of equalities

$$\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})\Theta[\mathbf{v}(\mathbf{q}-\mathbf{k})]}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}) =\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})\Theta(-\mathbf{vq})}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}) =\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vq})\Theta[\mathbf{v}(\mathbf{k}-\mathbf{q})]}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}), \quad \text{(C3)}$$

the first of which is obtained by the variable substitution $\mathbf{q}\to\mathbf{k}-\mathbf{q}$, then use of the property (A19). The second is then obtained by the substitution of $\mathbf{k}\to\mathbf{q}-\mathbf{k}$ and using the properties (A13) and (A17). By virtue of the identity

$$\Theta(\mathbf{vk})\Theta[\mathbf{v}(\mathbf{q}-\mathbf{k})]+\Theta(\mathbf{vq})\Theta[\mathbf{v}(\mathbf{k}-\mathbf{q})]+\Theta(\mathbf{vk})\Theta(-\mathbf{vq}) =\Theta(\mathbf{vk}),$$

Eq. (C3) yields

$$\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})\Theta[\mathbf{v}(\mathbf{q}-\mathbf{k})]}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}) =\frac{1}{3}\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}). \quad \text{(C4)}$$

The last step is to show that

$$\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}) =\int \frac{d\mathbf{q}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vq})}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}), \quad \text{(C5)}$$

which is done by three consecutive variable substitutions $\mathbf{k}\to\mathbf{q}-\mathbf{k}$, $\mathbf{q}\to\mathbf{k}-\mathbf{q}$, and ($\mathbf{k}\to-\mathbf{k}$, $\mathbf{q}\to-\mathbf{q}$) with application of the properties (A13), (A19), (A15), and (A17).

Combining Eqs. (C1),(C2), (C4), and (C5) we see that

$$\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vk})}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}) =\frac{1}{3}\int \frac{d\mathbf{k}d\mathbf{q}}{k^2q^2}\frac{\Theta(\mathbf{vq})}{|\mathbf{k}-\mathbf{q}|^2}\mathrm{Im}\chi_2^{\mathrm{TO}}(\mathbf{q},\mathbf{k},\mathbf{vq},\mathbf{vk}),$$

which proves the equivalence of Eqs. (39) and (41).

APPENDIX D: SOME INTEGRALS

In the case of forward scattering integrals (50) can be evaluated analytically. First we stretch the z axis along the common direction of vectors **v** and **k** and integrate over the polar angle of **q**. The results are the even functions of q and each integral can be evaluated by the residue theorem by closing the integration path in the appropriate complex half-plane. In the case of the integral A the singularities occur either at or off the real axis of q, depending on whether the incident energy is below or above $2\omega_p$, dramatically effecting the behavior of the quadratic contribution, as can be seen from Figs. 1 and 2. The results are

$$B(\omega_p)=\frac{6k\pi^3\omega_p}{v^2},$$

$$C(\omega_p)=\frac{k\pi^3(kv-4\omega_p)}{v^2}.$$

If $\mu v^2/2>2\omega_p$, then

$$A(2\omega_p)=\frac{k\pi^3}{v^2(k-v\mu)^2(kv-4\omega_p)}[16\omega_p^2(2k^2-4kv\mu+3v^2\mu^2)-k^2v^2(4k^2-9kv\mu+4v^2\mu^2)-4kv^2\mu(k+v\mu)\omega_p],$$

otherwise

$$A(2\omega_p)=\frac{2k^3\pi^3}{4\omega_p-kv}+\frac{k\pi^2}{v^2(kv-4\omega_p)p'^2}\times\{v^2[\pi-\mathrm{Arg}(q_1-q_0)-\mathrm{Arg}(q_1+q_0)]\times[k(k-2p)+4\mu\omega_p][k(p-2k)+4\mu\omega_p]-64\mathrm{Arg}(q_1)\omega_p^2p'^2+2(2k-3p)vp'\times\sqrt{\mu}(kv-4\omega_p)\sqrt{4\omega_p-v^2\mu}+32\pi\omega_p^2p'^2\},$$

where

$$q_0=\frac{\sqrt{k\mu(4\omega_p-kv)}}{\sqrt{v\mu-k}},$$

$$q_1=v\mu+i\sqrt{\mu}\sqrt{4\omega_p-v^2\mu},$$

and "Arg" is the argument of a complex number.

For an arbitrary angle of scattering we could not evaluate the integrals in a closed form, so the results in Fig. 2 are obtained by analytical integration over q, then by double numerical integration over the polar and azimuth angles. The results of the analytical and the numerical integrations have been checked to be identical for the forward scattering.

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