Nonretarded hydrodynamic-model calculation
of second-harmonic generation at a metal surface

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We show that within a hydrodynamic model the efficiency of second-harmonic generation at a vacuum-metal surface can be calculated to reasonable accuracy by a theory in which the speed of light is set to infinity. Both algebraic arguments and numerical results are presented. The simplifications allowed by an electrostatic calculation should also aid the evaluation of more sophisticated models.

I. INTRODUCTION

Two years ago Corvi and Schaich (CS) presented both formal and numerical results for the efficiency of second-harmonic generation in a simple reflection geometry.[1] Their theory uses a hydrodynamic model to describe the motion of free electrons at a smooth surface and the full set of Maxwell equations to determine the electromagnetic fields. Our aim in this paper is an explicit demonstration that the net result of their theory can be found from an analysis in which the speed of light c is at times formally allowed to be infinite. We will use the same hydrodynamic model for the constitutive relations, but will treat (up to a certain stage) the induced fields using only the electrostatic limit of Maxwell's equations.

The connection between the finite-c and infinite-c calculations is made via the phenomenological theory of Rudnick and Stern.[2,3] This theory writes the second-harmonic-generation efficiency for our model system as [Eqs. (CS-30), and (CS-31); see Ref. 4]

$$R = \frac{8\pi e^2}{m^2 \omega^2 c^3} |r|^2 , \tag{1}$$

where

$$r = \frac{\omega^2}{\tilde{\omega}^2}(\epsilon - 1)\alpha_0^2 \frac{Q\epsilon/2}{P_v\epsilon + P_T}$$
$$\times \left[a \left| \frac{Qc}{\omega} \right|^2 \frac{\epsilon}{\epsilon} - b\frac{p_T P_T c^2}{\omega^2\epsilon} + 2d\frac{\tilde{\omega}^2}{\tilde{\Omega}^2} \right] . \tag{2}$$

Here the dielectric functions are either at the first harmonic, $\epsilon = 1 - \omega_B^2/\tilde{\omega}^2$ with $\tilde{\omega}^2 = \omega(\omega + i/\tau)$, or at the second harmonic $\varepsilon = 1 - \omega_B^2/\tilde{\Omega}^2$ with $\tilde{\Omega}^2 = \Omega(\Omega + i/\tau)$ and $\Omega = 2\omega$. These functions allow one to find the normal components of the wave vectors of transverse fields far inside the metal, p_T and P_T, from the initial conditions of a wave of frequency ω incident at θ with respect to the surface normal. With $Q = (\omega/c)\sin\theta$ and $K = 2Q$, one has at the first harmonic $p_T = [(\omega^2/c^2)\epsilon - Q^2]^{1/2}$ and at

the second $P_T = [(\Omega^2/c^2)\varepsilon - K^2]^{1/2}$. The analogous components outside the metal are $p_v = (\omega/c)\cos\theta$ and $P_v = 2p_v$. From these definitions we can construct the first-order Fresnel transmission amplitude for the incident p wave, $\alpha_0 = 2p_v/(p_v\epsilon + p_T)$. Other terms needed in (1) and (2) are either fundamental constants, m, c, and $e < 0$, or bulk values of hydrodynamic parameters, the plasma frequency ω_B and the scattering rate $1/\tau$. Finally, we note the appearance in (2) of the phenomenological parameters a, b, and d. These are supposed to be functions of frequency alone (not θ) and require in general a separate microscopic calculation. CS showed that their (everywhere) finite-c, hydrodynamic calculation could be parametrized quite well by Eqs. (1) and (2). Indeed they extracted fitted values of a, b, and d and found that only a has a significant frequency dependence since $b \simeq -1$ and $d \simeq 1$.

We demonstrate here that one may reproduce their finite-c calculation of a using a theory in which $c \to \infty$; i.e., that an electrostatic calculation of a, together with Eqs. (1) and (2), allows one to accurately determine the second-harmonic generation efficiency. The importance of this conclusion is at the moment primarily conceptual, but it may have significant practical implications. Specifically for the hydrodynamic model, R can now be computed with c either always or only sometimes finite. Both computations require less than a second on a CDC Cyber 170/855, with the electrostatic calculation of a being roughly four times faster than the fully retarded one.

The real utility of our conclusion lies with future calculations that will employ electronic structure models more sophisticated than the hydrodynamic. For these the reduction in conceptual and computational effort provided by an electrostatic theory of a may be vital. We have in mind an extension of the recent calculations by Weber and Liebsch.[5] They use a jellium model treated within a local-density-functional approximation to find second-order induced charge densities at a metal surface. Their theory determines a in the double limit that $c \to \infty$ and $\omega \to 0$. The results we find here imply that only the $\omega \to 0$

limit need be removed in their theory to give it general applicability. Thus one may continue to focus on density response and to ignore transverse currents and fields. It is precisely this simplification that has led to the considerable advance in computational efficiency for surface corrections to the first-order optical response,[6-14] and we hope that its utility will be just as great for second-order properties.

In Sec. II we develop the equations necessary for an electrostatic calculation of a within our hydrodynamic model. The analysis is quite similar to that of CS. In the Appendix we compare their mathematical structure with those of the finite-c theory,[1] noting the behavior of the latter as $c \to \infty$. In Sec. III we show some explicit results that directly compare with those in CS and support our conclusion that an electrostatic calculation of a suffices.

II. BASIC EQUATIONS

Since the electrostatic theory only produces the a term in (2), not the complete R in (1), it has a somewhat different formal structure than the fully retarded theory. Hence we summarize its derivation before comparing in detail with the equations of CS. One studies the first- and second-harmonic response of a metal surface to an external field at frequency ω applied normal to the surface. With this perturbation and since the surface is presumed to be flat, the only spatial variation is with respect to x, the coordinate normal to the surface, and vectors may be written as scalars since they are all perpendicular to the surface. The basic variables we study are the total electric field E and the polarization of the metal P. From the latter we can obtain the induced charge density $\rho = -\partial P/\partial x$ and the current density $j = \partial P/\partial t$. As did CS, we expand these quantities in a series of orders and focus on the spatial dependence of the complex amplitudes such as the $P_j(x)$ for the polarization;

$$P(x,t) = 2\,\mathrm{Re}[P_1(x)e^{-i\omega t} + P_2(x)e^{-2i\omega t} + \cdots]\,, \qquad (3)$$

where Re denotes "real part of."

The a parameter is found from the integrated weight of P_2:

$$\int dx\, P_2(x) = \frac{a}{4\rho_B}\left[\frac{E_1^{(\mathrm{out})}}{4\pi}\left[\frac{1-\epsilon}{\epsilon}\right]\right]^2\,, \qquad (4)$$

where $E_1^{(\mathrm{out})}$ is the (constant) value of the total electric field at the first harmonic outside the metal and $\rho_B < 0$ is the bulk equilibrium electron charge density. The metal lies in $x > 0$ and we need to require that $P_2(x \to \infty) \to 0$ in order that the integral be well defined. This is equivalent to the constraint that the total induced charge at the second harmonic is zero for the single surface problem.

Our basic method of finding $P_2(x)$ is the same as in CS. We imagine that the equilibrium electron charge-density profile of the metal can be represented by a sequence of steps of constant height, say, ρ_0. Then within each step we expand the full solution in terms of "partial waves," which individually satisfy the equations of motion there. Finally, boundary conditions are imposed in order to match these expansions between successive steps. This

scheme is applied twice, beginning with the first-harmonic equations whose solution is used to generate driving terms for the second-harmonic equations.

The formal derivation of the equations of motion at each harmonic is essentially identical to that in CS, so we simply list them here and note their partial-wave solutions. At the first harmonic one has

$$\left[-\tilde{\omega}^2 - \beta_0^2\frac{d^2}{dx^2}\right]P_1(x) = \frac{\omega_0^2}{4\pi}E_1(x)\,, \qquad (5)$$

where β_0 is the local velocity parameter and ω_0 the local plasma frequency. At each internal interface we require continuity of P_1, E_1, and $\beta_0^2\rho_1/\rho_0$. At the external interface (i.e., at $x=0$, where vacuum starts), we require E_1 to be continuous and P_1 to vanish. The partial wave solutions of (5) are of two kinds: either constant, denoted (C), or spatial varying, denoted (L). For the former

$$4\pi P_1^{(C)} = -\frac{\omega_0^2}{\tilde{\omega}^2}E_1^{(C)} \qquad (6)$$

while for the latter

$$P_1^{(L)}(x) \propto e^{\pm ip_L x} \qquad (7)$$

with

$$p_L^2 = (\tilde{\omega}^2 - \omega_0^2)/\beta_0^2\,, \qquad (8)$$

and

$$4\pi P_1^{(L)}(x) = -E_1^{(L)}(x)\,. \qquad (9)$$

The L subscript denotes "longitudinal," since only these solutions have an associated charge density. The constant solutions are the remnant of the transverse waves that appear in the finite-c theory.[1] The boundary conditions on E_1 and P_1 imply that $(1 - \omega_0^2/\tilde{\omega}^2)E_1^{(C)}$ is the same everywhere, which merely describes the classical screening of the external field.

At the second harmonic one has

$$\left[-\tilde{\Omega}^2 - \beta_0^2\frac{d^2}{dx^2}\right]P_2(x) = \frac{\omega_0^2}{4\pi}E_2(x) + S(x)\,, \qquad (10)$$

where the driving terms are determined by the first-order solution;

$$S(x) = -\frac{1}{2\rho_0}\frac{d}{dx}(j_1)^2 + \frac{\rho_1}{\rho_0}\left[\frac{\omega_0^2}{2\pi}E_1 + \frac{5}{3}\beta_0^2\frac{d^2}{dx^2}P_1 - \frac{1}{\tau}j_1\right]\,. \qquad (11)$$

Equation (14) is the analog of Eq. (CS-14), with the omission of the magnetic field contribution. It is worth remarking that $S(x)$ has no position-independent contribution, so it is nonzero only close to the surface because the longitudinal, first-order solutions are all chosen to decay as $x \to \infty$. Within a general step $S(x)$ has contributions that vary as $e^{+iQ_l x}$, where Q_l has four possible values: $\pm p_L$ or $\pm 2p_L$. The partial-wave solution driven, (D), by $u_l e^{iQ_l x}$ is

$$P_2^{(D,l)}(x) = \frac{u_l/\beta_0^2}{Q_l^2 - P_L^2} e^{iQ_l x} \tag{12}$$

with

$$P_L^2 = (\tilde{\Omega}^2 - \omega_0^2)/\beta_0^2 , \tag{13}$$

and

$$4\pi P_2^{(D,l)}(x) = -E_2^{(D,l)}(x) . \tag{14}$$

We must also include solutions of (10) when S is zero. As in the first-harmonic case these are either constant or spatially varying. With an analogous notation we have

$$4\pi P_2^{(C)} = -\frac{\omega_0^2}{\tilde{\Omega}^2} E_2^{(C)} \tag{15}$$

for the former and

$$P_2^{(L)}(x) \propto e^{\pm iP_L x} \tag{16}$$

with

$$4\pi P_2^{(L)}(x) = -E_2^{(L)}(x) \tag{17}$$

for the latter.

At each internal interface we require continuity of P_2, E_2, and

$$\frac{1}{2}v_1^2 + \frac{\beta_0^2}{\rho_0}\left[\rho_2 - \frac{1}{6}\frac{\rho_1^2}{\rho_0}\right] ,$$

where $v_1 = j_1/\rho_0$; while at the external interface we require both E_2 and P_2 to vanish. The continuity of P_2 and E_2 together with (14), (15), and (17) imply that $(1 - \omega_0^2/\tilde{\Omega}^2)E_2^{(C)}$ is the same everywhere. The additional assumption that E_2 vanishes outside implies that no constant partial wave is allowed anywhere. As noted earlier this is done so that a in (4) is unambiguous.

We have now set the basic equations for the nonretarded calculation of a. Their evaluation on a computer is straightforward. For an n-step model one needs to work with matrices of dimension $2n - 1$. In contrast, the fully retarded, n-step model calculation used matrices of di-

mension $4n - 1$. The difference arises because there are transverse waves in the retarded case moving along $\pm x$ within each step while in the electrostatic case the constant fields can be separately found and removed.

In the Appendix we compare the two sets of equations in greater detail. This explicit analysis complements the formal arguments given earlier by Sipe et al.[15,16] that an electrostatic theory of a can suffice. In the next section we furthermore present specific electrostatic solutions whose implications may be numerically compared with those of the fully retarded theory.

III. ILLUSTRATIVE EXAMPLES

Begin with a single-step model; i.e., bulk metal in $x > 0$ and vacuum in $x < 0$. The electrostatic problem then simplifies enough that we can present the explicit solution. At first order

$$4\pi P_1(x) = -E_1^{(out)} \times \begin{cases} 0, & x < 0 \\ \dfrac{\omega_B^2}{\tilde{\omega}^2}(1 - e^{iP_L x})/\epsilon, & 0 < x \end{cases} \tag{18}$$

$$E_1(x) = E_1^{(out)} \times \begin{cases} 1, & x < 0 \\ \left[1 - \dfrac{\omega_B^2}{\tilde{\omega}^2}e^{iP_L x}\right]\Big/\epsilon, & 0 < x . \end{cases} \tag{19}$$

Substituting into (11) yields for the driving terms in the metal

$$S(x) = \left[\frac{E_1^{(out)}}{4\pi}\left[\frac{1-\epsilon}{\epsilon}\right]\right]^2 \frac{iP_L}{\rho_B}(v_1 e^{iP_L x} + v_2 e^{2iP_L x}) , \tag{20}$$

where

$$v_1 = -(3\omega^2 + i\omega/\tau) , \tag{21}$$

$$v_2 = (\omega_B^2 + 8\omega^2 + 2i\omega/\tau)/3 . \tag{22}$$

The second-order solution is, for $x > 0$,

$$P_2(x) = \left[\frac{E_1^{(out)}}{4\pi}\left[\frac{1-\epsilon}{\epsilon}\right]\right]^2 \frac{iP_L}{\beta_B^2 \rho_B}\left[\frac{v_1}{p_L^2 - P_L^2}(e^{iP_L x} - e^{iP_L x}) + \frac{v_2}{(2p_L)^2 - P_L^2}(e^{2iP_L x} - e^{iP_L x})\right] \tag{23}$$

with $E_2 = -4\pi P_2$ there. Both vanish for $x < 0$. Finally the definition (4) yields

$$a = \frac{4}{\beta_B^2 P_L}\left[\frac{v_1}{p_L + P_L} + \frac{v_2/2}{2p_L + P_L}\right] , \tag{24}$$

where Eqs. (8) and (13) are to be evaluated with bulk parameters, $\beta_0 = \beta_B$ and $\omega_0 = \omega_B$.

With this formula we can reproduce the plot of the fitted a shown in Fig. 2 of Ref. 1. If we further set $b = -1$ and $d = 1$ and use (1) and (2), we also duplicate the R shown there. Note that the primary structure in

this a comes from the P_L^{-1} factor, the sharpness of which is controlled by the choice of $1/\tau$. The limiting values of a are material independent:

$$a \to \begin{cases} -\frac{2}{9}, & \omega \to 0 \\ -\frac{4}{3}, & \omega \to \infty . \end{cases} \tag{25}$$

If we now add a surface layer to our system to form a double-step model, the algebra becomes sufficiently involved that we only present numerical results. The parameters are chosen identical to those of the double-step model of CS, illustrated in their Figs. 5 and 6. The model

simulates Al using a surface ledge of width 4 Å and density $\rho_0 = 0.7\rho_B$.[17] The scattering rate satisfies everywhere $\omega_B \tau = 10$.

In Fig. 1 we show an analogue of Fig. 6 from CS. The electrostatic a reproduces all the structure found in the fully retarded calculation. The agreement is not perfect, but is typically better than a few percent. We checked that the retarded answers become closer to the electrostatic results as we (artificially) increase the speed of light. The rate of convergence is consistent with the arguments in the Appendix.

We have compared results for several other sets of parameter choices. The agreement remains good. We also examined the spatial variation of the fields, which is integrated over to form a. The differences in these profiles occasionally exceed a few percent, especially when the induced longitudinal fields are weak. However, an increase in c by about a factor of 10 restores agreement. Hence we remain confident that strong structure in a can be efficiently calculated with adequate accuracy by setting the speed of light to infinity. We feel that this conclusion is more generally valid than the hydrodynamic model used here, because the important corrections to this model do not involve retardation effects.
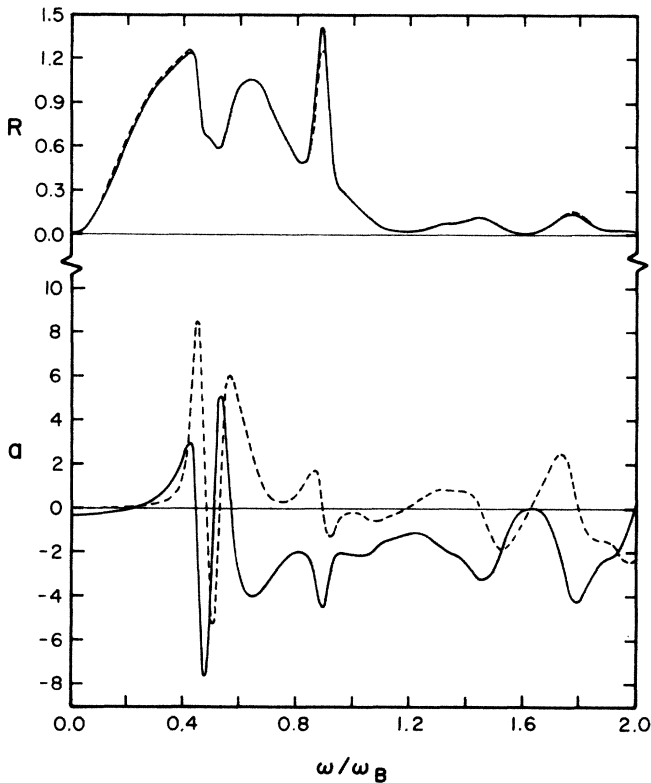

FIG. 1. Frequency dependence of the second-harmonic generation parameter a that results from an electrostatic calculation for a double-step model of Al. In the lower panel the real (imaginary) part of a is a solid (dashed) line. The upper panel shows R in units of 10^{-20} cm^2/W. The solid (dashed) line is for the nonretarded (retarded) calculation. Other model parameters are given in the text.

APPENDIX

Here we briefly outline algebraic reasons why the finite-c theory[1] should yield the same a in the limit $c/\beta_0 \rightarrow \infty$ as the electrostatic theory described in Sec. II. The key point is that the normal component of wave vectors of longitudinal fields,[18] p_L and P_L, become in this limit much larger than any other wave-vector components such as Q, K, p_T, p_v, P_T, or P_v.

Consider the first-order polarization in the retarded case. Its partial-wave decomposition within any step has terms of both transverse,

$$\alpha_1^{(T,\pm)}(Q, \mp p_T, 0)e^{\pm ip_T x + i\overline{Q}\cdot\overline{X}} ,$$

and longitudinal,

$$\alpha_1^{(L,\pm)}(\pm p_L, Q, 0)e^{\pm ip_L x + i\overline{Q}\cdot\overline{X}} ,$$

form. Here the two-dimensional vectors $\overline{Q}$ and $\overline{X}$ lie in the surface plane and the triplet of numbers denote the vector components along the surface normal $\hat{x}$, along $\hat{Q}$, and along $\hat{t} = \hat{x} \times \hat{Q}$, respectively.[1] The requirements of continuous $\hat{x} \cdot \mathbf{P}_1$, $\hat{x} \cdot \mathbf{E}_1$, $\hat{Q} \cdot \mathbf{E}_1$, and $\beta_0^2 \rho_1 / \rho_0$ make

$$Q\alpha_1^{(T,\pm)} \sim p_L \alpha_1^{(L,\pm)} , \tag{A1}$$

where $\sim$ denotes "of the same magnitude in the limit of large c/β_0." This implies that $\rho_1 = -\nabla \cdot \mathbf{P}_1$ will be dominated by $-(\partial/\partial x)(\hat{x} \cdot \mathbf{P}_1)$ and that the full driving term may be approximated by

$$\mathbf{S} = \frac{\omega_0^2}{4\pi\rho_0 c}(\mathbf{j}_1 \times \mathbf{B}_1) - \frac{1}{\rho_0}(\mathbf{j}_1 \cdot \nabla)\mathbf{j}_1$$

$$+ \frac{\rho_1}{\rho_0}\left[\frac{\omega_0^2}{2\pi}\mathbf{E}_1 - \frac{5}{3}\beta_0^2\nabla\rho_1 - \frac{1}{\tau}\mathbf{j}_1\right]$$

$$\simeq \hat{x}\frac{\omega^2}{2\rho_0}\frac{\partial}{\partial x}(\hat{x}\cdot\mathbf{P}_1)^2$$

$$+ \frac{\rho_1}{\rho_0}\left[\frac{\omega_0^2}{2\pi}\mathbf{E}_1 + \hat{x}\frac{5}{3}\beta_0^2\frac{\partial^2}{\partial x^2}(\hat{x}\cdot\mathbf{P}_1) + \frac{i\omega}{\tau}\mathbf{P}_1\right] ,$$

$$\tag{A2}$$

if one considers only its effect near the surface. This last constraint is necessary in order to suppress the small but long-range contribution to $\mathbf{S}$ from the magnetic field, $\mathbf{B}$.[19] This contribution leads to the d term in (2), but is irrelevant for the calculation of a and b, which require only the short-range part of the polarization field.[19]

Now consider the second-order polarization in the retarded case. Within any step its driving terms are dominated by functions of the form

$$u_t(K, -P_t, 0)e^{iP_t x + i\overline{K}\cdot\overline{X}}$$

or

$$u_l(P_l, K, 0)e^{iP_l x + i\overline{K}\cdot\overline{X}} \ ,$$

where $\overline{K} = 2\overline{Q}$ and $P_t \sim P_l \sim p_L$. If we choose the incident first-order field $E_1 \sim 1$, then $u_t \sim u_l \sim 1$, too. The solutions of the analogue of (10) [Eq. (CS-13)], are quite sensitive to the polarization of the driving term. For the longitudinal case l one finds [cf. (12)–(14)]

$$\mathbf{P}_2^{(D,l)} \simeq \frac{u_l/\beta_0^2}{P_l^2 - P_L^2}(P_l, K, 0)e^{iP_l x + i\overline{K}\cdot\overline{X}} \tag{A3}$$

with

$$\mathbf{E}_2^{(D,l)} = -4\pi\mathbf{P}_2^{(D,l)} \ ; \tag{A4}$$

while for the transverse case, which does not arise in the electrostatic theory,

$$\mathbf{P}_2^{(D,t)} \sim -\frac{u_t}{\widetilde{\Omega}^2}(K, -P_t, 0)e^{iP_t x + i\overline{K}\cdot\overline{X}} \tag{A5}$$

and

$$\mathbf{E}_2^{(D,t)} \sim -4\pi\frac{\widetilde{\Omega}^2}{\omega_0^2}\mathbf{P}_2^{(D,t)}/(c/\beta_0)^2 \ . \tag{A6}$$

We must augment the above with partial-wave solutions of the homogeneous second-order equations. These are either transverse,

$$\alpha_2^{(T,\pm)}(K, \mp P_T, 0)e^{\pm iP_T x + i\overline{K}\cdot\overline{X}} \ ,$$

or longitudinal,[18]

$$\alpha_2^{(L,\pm)}(\pm P_L, K, 0)e^{\pm iP_L x + i\overline{K}\cdot\overline{X}} \ ,$$

and the α's are to be determined by matching boundary conditions. Provided $1/\tau$ is the same everywhere (see below), we can satisfy the continuity constraints on

$$\hat{\mathbf{x}}\cdot\mathbf{P}_2$$

and

$$\tfrac{1}{2}(\mathbf{v}_1)^2 + \frac{\beta_0^2}{\rho_0}\left[\rho_2 - \frac{1}{6}\frac{\rho_1^2}{\rho_0}\right]$$

by focusing on longitudinal waves alone. The driving terms make $\alpha_2^{(L,\pm)} \sim 1$. In fact, the equations satisfied by the $\alpha_2^{(L,\pm)}$ are as $c/\beta_0 \rightarrow \infty$ isomorphic to the electrostatic ones. The other continuity requirements of $\hat{\mathbf{Q}}\cdot\mathbf{E}_2$ and $\hat{\mathbf{x}}\cdot(\mathbf{E}_2 + 4\pi\mathbf{P}_2) = \hat{\mathbf{x}}\cdot(\mathbf{E}_2^{(T)} + 4\pi\mathbf{P}_2^{(T)})$ do not involve large

terms (e.g., $\hat{\mathbf{x}}\cdot\mathbf{P}_2^{(L)}$ or $\hat{\mathbf{Q}}\cdot\mathbf{P}_2^{(T)}$) and hence yield consistently $\alpha_2^{(T,\pm)} \sim 1$.

These arguments lead to the conclusion that the short-range normal component of the second-order polarization is dominated (to within corrections of order β_0/c) by the longitudinal field calculated from electrostatics using $E_1^{(out)} = \hat{\mathbf{x}}\cdot\mathbf{E}_1^{(out)}$. The numerical results for a in Sec. III support this argument. For the transverse component matters are even simpler since the dominant part of $\hat{\mathbf{Q}}\cdot\mathbf{E}_1 \simeq -4\pi(\widetilde{\omega}^2/\omega_0^2)\hat{\mathbf{Q}}\cdot\mathbf{P}_1$ is constant on the short-range scale. Hence[20]

$$\int dx\,\hat{\mathbf{Q}}\cdot\mathbf{P}_2 \simeq -\int dx\,\hat{\mathbf{Q}}\cdot\mathbf{S}/\widetilde{\Omega}^2$$

$$\simeq -(\hat{\mathbf{Q}}\cdot\mathbf{E}_1)\int dx\left[\frac{\omega_0^2}{8\pi\rho_0}\right]\rho_1/\widetilde{\omega}^2 \ , \tag{A7}$$

but $\omega_0^2/8\pi\rho_0 = e/2m$ and

$$\int dx\,\rho_1 \simeq \frac{1}{4\pi}(\hat{\mathbf{x}}\cdot\mathbf{E}_1^{(out)})\left[\frac{1-\epsilon}{\epsilon}\right] \ . \tag{A8}$$

Together (A7) and (8) imply that

$$b \simeq -1 \ . \tag{A9}$$

Finally we return to the remark that $1/\tau$ must be the same in each step for the above to hold. This extra condition arises from the need to make equivalent as $c/\beta_0 \rightarrow \infty$ the requirements at internal interfaces of continuity of

$$\tfrac{1}{2}(\mathbf{v}_1)^2 + \frac{\beta_0^2}{\rho_0}\left[\rho_2 - \frac{1}{6}\frac{\rho_1^2}{\rho_0}\right]$$

and

$$\tfrac{1}{2}v_1^2 + \frac{\beta_0^2}{\rho_0}\left[\rho_1 - \frac{1}{6}\frac{\rho_1^2}{\rho_0}\right]$$

for the retarded and electrostatic theories, respectively. If $1/\tau$ is continuous through an interface, then $\hat{\mathbf{Q}}\cdot\mathbf{v}_1$ is also continuous as $c/\beta_0 \rightarrow \infty$ because it is dominated by transverse waves which satisfy

$$\hat{\mathbf{Q}}\cdot\mathbf{v}_1^{(T)} = i\frac{\omega_0^2}{4\pi\rho_0}\frac{\hat{\mathbf{Q}}\cdot\mathbf{E}_1^{(T)}}{\omega + i/\tau} \tag{A10}$$

and because we already require $\hat{\mathbf{Q}}\cdot\mathbf{E}_1$ to be continuous. We checked numerically that allowing $1/\tau$ to be different in different steps created noticeable differences in R between the electrostatic and retarded theories. These differences could not be removed by increasing c.

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[18]The definitions of p_L and P_L in Ref. 1 differ from those used here by terms of relative order β_0^2/c^2, which we ignore.

[19]Here "long range" means on the scale of a transverse wavelength, while "short range" means comparable to a longitudinal wavelength, which is typically the same size as the depth of surface diffuseness.

[20]The integral runs over a short range near the surface.