Bulk and surface dielectric response of a superlattice with an arbitrary varying dielectric function: A general analytical solution in local theory in the long-wave limit

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We present an analytical solution for the dielectric response of infinite and semi-infinite superlattices with the constituent dielectric function $\epsilon(\omega, z)$ being an arbitrary periodic function of one coordinate z. The long-wave limit in the local theory is used. All results are expressed in terms of the two bulk quantities, namely, the average over one period of the functions $\epsilon(\omega, z)$ and $1/\epsilon(\omega, z)$. The damping of the bulk and the surface plasmon modes specific for superlattices with a continuously varying constituent dielectric function is obtained and discussed. Our theory provides deeper insight into the role of the local-field effects in the dielectric response of a superlattice.

I. INTRODUCTION

Within the last two decades, there has been a large amount of work devoted to the superlattices, which are systems consisting of alternating layers with different dielectric properties. Using the hydrodynamical approximation, Fetter[1] obtained the spectrum of collective excitations in the layered electron gas, which is an infinite periodic array of sheets of two-dimensional electron gas. Giuliani and Quinn[2] obtained the quantum-mechanical solution for the semi-infinite variant of this model. Giuliani, Quinn, and Wallis[3] derived the dispersion relation for plasmons in an infinite metallic superlattice consisting of layers of finite width. Camley and Mills[4] solved this problem for infinite and semi-infinite superlattices with arbitrary dielectric functions of the layers. In the latter two works the local approximation was used.

Since then, the theory of collective excitations in layered systems has included many of the more fine effects: the finiteness of a superlattice,[5] the intrinsic damping of the superlattice constituents and the retardation,[6] the coupling of two adjoining superlattices[7] (for more references see, for example, Ref. 8).

However different these models are with respect to the studied superlattices or in the approximations applied, they have one important feature in common: the superlattices are assumed to consist of alternating layers of two (or more) completely distinct materials with their own bulk dielectric functions. The boundaries between layers are assumed to be abrupt and the constituents of the superlattices are suggested to pertain to their bulk dielectric function up to the layers's boundaries.

We find it interesting to consider a superlattice with a continuously varying dielectric function of the constituents. The most evident reason for this is that the interfaces in real superlattices may not be abrupt and it is important to account for the intermediate regions. Another reason, as we shall show, is that the superlattices with a continuously varying dielectric function demonstrate an effect not present in the superlattices with abrupt layers: the former have a damping of collective excitations which does not originate from the imaginary part of the constituent dielectric function but is the property of the superlattice as a whole.

In this paper, we present the analytical solutions for the dielectric response and the eigenmodes of collective excitations of infinite and semi-infinite superlattices constituted by a dielectric function $\epsilon(\omega, z)$, which is an arbitrary periodic function of coordinate z. These solutions are obtained in the local theory and in the long-wave limit. We show that the bulk macroscopic dielectric function of the superlattice as a whole, the surface energy loss function, and the excitation spectra in both the infinite and the semi-infinite cases depend on only two bulk quantities: the average over the period of the functions $\epsilon(\omega, z)$ and $1/\epsilon(\omega, z)$.

The spectrum of the bulk modes in the long-wave limit has a directional spatial dispersion, which is the property of the superlattice as a whole, and reflects the anisotropy of the system.

In this paper, we show that for a superlattice the conventional expression for the surface energy loss function of a homogeneous medium $L_s(\omega) = -\text{Im}\, 1/[1 + \epsilon(\omega)]$ (Ref. 9) must be replaced by

$$L_s(\omega) = -\text{Im}\, \frac{1}{1 + \sqrt{\epsilon_{\parallel}(\omega)\epsilon_{\perp}(\omega)}},$$

where $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are the parallel and the normal to the surface components of the dielectric tensor of the superlattice as a whole.

We discuss the role of the local-field effects in the theory of the dielectric response of superlattices and we show that, in contrast to the dielectric theory of the crystalline solids,[10] the neglect of these effects for superlattices is totally unacceptable, since it is equivalent to the replacement of a superlattice with a homogeneous dielectric medium.

In this work we demonstrate that the technique of inverting the dielectric matrix used in the dielectric theory of crystalline solids can be successfully applied in the theory of superlattices, which is the alternative approach to the direct solving of the Maxwell's equations or the transfer matrix method.

Let us note that, although the long-wave limit is a specific case in the theory of the dielectric response of superlattices, it is a very important one. For example, the optical spectra of superlattices are characterized by the long-wave limit of the dielectric function of a superlattice as a whole. Similarly, for the fast incident electrons, the energy loss spectra are mostly defined by the long-wave behavior of the dielectric response.

Apart from these, there is one philosophical reason why the long-wave properties of semi-infinite superlattices are especially important. Only in the long-wave limit the dielectric response and eigenmodes of semi-infinite superlattices do not depend on the spot within the period where the surface is cut, so that we can speak of the response of a semi-infinite superlattice without specifying the detailed structure of its surface.

Since our results are obtained for an arbitrary periodic dielectric function, they are also applicable for the steplike dielectric functions, characterizing the superlattices with distinct constituents. For this case, we present the comparison of our results with the long-wave limit of the results known from the literature, and we obtain an agreement.

II. DIELECTRIC RESPONSE OF INFINITE SUPERLATTICE

A. The range of applicability of the local approximation in the theory of the dielectric response of superlattices

To outline the range of applicability of the local approximation in the theory of superlattices, let us first consider this question for more conventional superlattices with distinct constituents.

The selection of an approximation to investigate a solid superlattice depends on the relations between the three characteristic scales of the problem: the length of the order of the periods of the crystalline lattice of the superlattice constituents a, the period of the superlattice c, and the wavelength λ of the electromagnetic wave propagating in the superlattice. We can regard the following cases.

(1) c is of the same order as a. In this case the layers of the superlattice are not of sufficient width to treat them macroscopically. A quantum-mechanical approach must be used.

(2) $c \gg a$, λ is of the same order as a. The layers of the superlattice constituents can be considered macroscopically. The response in an individual layer can be described by macroscopic nonlocal frequency and a wave-vector-dependent bulk dielectric function $\epsilon(\omega, \mathbf{q})$ of the corresponding infinite material, which can be calculated or taken from an independent experiment. Then the electromagnetic problem of coupling the excitations at the interfaces of the superlattice must be solved.

(3) $c \gg a$, $\lambda \gg a$. In this case we can describe the dielectric response of an individual layer of the superlattice macroscopically by the frequency-dependent wave-vector-independent dielectric function $\epsilon(\omega)$, which is the local approximation. Since $\epsilon(\omega)$ does not depend on $\mathbf{q}$, we can write $\mathbf{D}(\mathbf{r}, \omega) = \epsilon(\omega)\mathbf{E}(\mathbf{r}, \omega)$ in each individual layer of the superlattice, where $\mathbf{D}$ is the dielectric displacement and $\mathbf{E}$ is the electric field. As in case 2, the electromagnetic problem of coupling the excitations at the interfaces must be solved, which, however, in this case is far more simple. It must be noted that although in this case the individual layers are described by the wave-vector-independent local dielectric function, the dielectric function and the excitation spectrum of the superlattice as a whole obtained in this approximation are wave-vector dependent and consequently nonlocal.[3,4] Let us also note that however simple this approximation appears, it is the one in which sophisticated spectra of excitations of superlattices were obtained (see, for example, Refs. 3, 4).

To apply the local approximation to superlattices with continuously varying dielectric function of the constituents, we must add one more condition to the conditions of case 3. It is $|d\epsilon(\omega, z)/dz| \ll |\epsilon(\omega, z)/a|$, which means that the dielectric function varies slowly over the atomic scale. Of course, unless this last condition is satisfied, the dielectric function of the superlattice constituents has no meaning at all.

The present work treats case 3. This means that we restrict our consideration to superlattices (metallic, semiconductor, dielectric, or mixed) with periods large compared with the atomic scale. While we study the excitations with wavelengths large compared with the period of the superlattice, the second of the conditions of case 3 is satisfied automatically. This case embraces a large family of physically interesting superlattices[4] with periods in the range of 100–5000 Å.

We neglect the retardation effects throughout the paper.

B. Microscopic dielectric matrix of infinite periodic system in local theory

It is the general practice to seek the dielectric response of superlattices with distinct constituents by solving the equations of the electromagnetic field in individual layers, then applying the boundary conditions at the layers's interfaces. Recently this technique was generalized to include in closed form any number of surfaces and interfaces.[11] However, it is evident that for a superlattice with continuously varying dielectric function no boundary conditions exist, and one must solve Maxwell's equations together with the material equations for the varying dielectric function. In the local theory these equations are

$$\nabla \cdot \mathbf{D}(\mathbf{r}, \omega) = 4\pi\rho_{\text{ext}}(\mathbf{r}, \omega), \tag{1}$$

$$\mathbf{D}(\mathbf{r}, \omega) = \epsilon(\omega, \mathbf{r})\mathbf{E}(\mathbf{r}, \omega), \tag{2}$$

where ρ_{ext} is the external charge density and $\epsilon(\omega, \mathbf{r})$ is the local dielectric function within a period of the superlattice. We assume that the medium is locally isotropic, so that $\epsilon(\omega, \mathbf{r})$ is scalar.

If the retardation effects are neglected, then we can describe the total and the external fields by their scalar potentials ϕ and ϕ_{ext}. Equations (1) and (2) then give

$$\nabla[\epsilon(\mathbf{r})\nabla\phi(\mathbf{r})] = \nabla^2\phi_{\text{ext}}(\mathbf{r}), \tag{3}$$

where from here on the ω dependence will be implied.

It is known[12,13] that, if the system under study is periodic, then in wave-vector representation the equation holds

$$\phi_{\text{ext}}(\mathbf{G}+\mathbf{q}) = \sum_{\mathbf{G}'} \epsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q})\ \phi(\mathbf{G}'+\mathbf{q}),$$

where $\epsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q})$ is the microscopic dielectric matrix of the system, $\mathbf{G}$ and $\mathbf{G}'$ are the reciprocal vectors of the periodic lattice, and $\mathbf{q}$ is the wave vector in the first Brillouin zone. It is also known[12,13] that

$$\epsilon_M(\mathbf{q}) = \frac{1}{\epsilon_{00}^{-1}(\mathbf{q})}, \tag{4}$$

where $\epsilon_M(\mathbf{q})$ is the macroscopic dielectric function of the system as a whole. It is seen from Eq. (4) that to obtain the macroscopic dielectric function, we must invert the microscopic dielectric matrix $\epsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q})$.

Equation (4) includes the local-field effects, which are the short-wave response to the long-wave perturbation. The approximation

$$\epsilon(\mathbf{q}) \approx \epsilon_{00}(\mathbf{q}) \tag{5}$$

is known as the one without the local-field effects.[10] Equation (4) is quite general for periodic systems and it is applicable for crystalline solids with periods of a few Å as well as for superlattices with periods of several thousands Å. In this work we apply the general formalism of inverting the microscopic dielectric matrix to a superlattice with an arbitrary periodic dielectric function of the constituents. This procedure is equivalent to the solving of Eq. (3) in the real space.

The explicit form of the microscopic dielectric matrix in local approximation can be obtained from Eq. (3) by going over into the wave-vector representation. Then we have

$$\epsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q}) = \frac{(\mathbf{G}+\mathbf{q})(\mathbf{G}'+\mathbf{q})}{(\mathbf{G}+\mathbf{q})^2}\epsilon(\mathbf{G}-\mathbf{G}'), \tag{6}$$

where $\epsilon(\mathbf{G})$ on the right-hand side is the Fourier transform of $\epsilon(\mathbf{r})$.

Let us note that the term "microscopic," as it is used in this paper, applies to the quantities of the individual periods of the superlattice, while "macroscopic" pertains to the lengths of a number of the periods of a superlattice. In this sense Eq. (2) is microscopic, although in the atomic scale it is, of course, macroscopic.

It can be seen from Eq. (6) that in the local approximation $\epsilon_{\mathbf{G}\mathbf{G}}(\mathbf{q}) = \epsilon(\mathbf{0})$ = *an average over the period of the constituent dielectric function* and it does not depend on $\mathbf{G}$ or $\mathbf{q}$. This rather striking fact must be interpreted as follows: in the local approximation the difference between the microscopic dielectric matrix of a superlattice and a homogeneous medium with the dielectric function equal to the average dielectric function of the superlattice constituents lies in the nondiagonal elements. If the strict procedure of inverting this matrix is carried out, then the resulting macroscopic dielectric function (4) of the superlattice will have the $\mathbf{q}$ dependence, which will be the same as that obtained by the solving of Maxwell's equations. *A priori* this statement follows from the equivalence of the Eqs. (1) and (2) and Eq. (6). *A posteriori* we show that in Sec. IV by comparing our results with these of the previous theories.

C. Inversion of the microscopic dielectric matrix of infinite superlattice

Let us write down the explicit form of the microscopic dielectric matrix for a superlattice constituted by periodic dielectric function $\epsilon(\omega, z)$, which is homogeneous in the XY plane. By Eq. (6), we have

$$\epsilon_{GG'}(q_z, q_\parallel) = \frac{(G+q_z)(G'+q_z)+q_\parallel^2}{(G+q_z)^2+q_\parallel^2}\epsilon(G-G'), \tag{7}$$

where $\epsilon(G)$ on the right-hand side is the Fourier transform of $\epsilon(z)$, $G = 2\pi n/c$, $n = 0, \pm1, \ldots$, and c is the period of the superlattice.

Let us invert Eq. (7) in the long-wave limit for an arbitrary angle θ between $\mathbf{q}$ and the superlattice axis z. To do it, let us introduce the matrix U with elements

$$U_{GG'} = [q_\parallel - i(G+q_z)]\delta_{GG'}, \tag{8}$$

where $\delta_{GG'}$ is Kronecker's symbol.

Then we can write

$$\begin{aligned}\epsilon'_{GG'}(q_z, q_\parallel) &\equiv (U\ \epsilon\ U^{-1})_{GG'}(q_z, q_\parallel) \\ &= \epsilon(G-G') \\ &\quad -iq_\parallel \frac{G-G'}{[q_\parallel + i(G+q_z)][q_\parallel - i(G'+q_z)]} \\ &\quad \times\epsilon(G-G').\end{aligned} \tag{9}$$

If q tends to zero, then it is seen from Eq. (9) that

$$\begin{aligned}\epsilon'_{GG'}(q_z, q_\parallel) &= \epsilon(G-G') - \epsilon(G-G') \\ &\quad \times \begin{cases} 0,\ G\neq 0, G'\neq 0 \text{ or } & G=0, G'=0 \\ \tan\theta/(\tan\theta - i), & G\neq 0, G'=0 \\ \tan\theta/(\tan\theta + i), & G=0, G'\neq 0 \end{cases}\end{aligned} \tag{10}$$

A matrix with the structure of Eq. (10) can be easily inverted (for details see Ref. 14). Then we can obtain with help of Eq. (8)

$$\epsilon^{-1}_{GG'}(\theta) = [U^{-1}\,(\epsilon')^{-1}\,U]_{GG'}$$
$$= \lim_{q\to 0}\left[\frac{q\exp(i\theta)+G'}{q\exp(i\theta)+G}\right]\frac{\sin\theta}{\Delta}\left\{\left(\frac{1}{\epsilon}\right)(G-G') + \sin\theta\left[\frac{1}{\epsilon_\perp}\delta_{G0}\delta_{0G'} - \epsilon_\parallel\left(\frac{1}{\epsilon}\right)(G)\left(\frac{1}{\epsilon}\right)(-G')\right]\right.$$
$$\left. +i\cos\theta\left[\delta_{G0}\left(\frac{1}{\epsilon}\right)(-G') - \delta_{0G'}\left(\frac{1}{\epsilon}\right)(G)\right]\right\}, \tag{11}$$

where $(1/\epsilon)\,(G)$ is the Fourier transform of $1/\epsilon(z)$,

$$\Delta = \cos^2\theta + \sin^2\theta\frac{\epsilon_\parallel}{\epsilon_\perp},$$
$$\epsilon_\parallel = \overline{\epsilon(z)},$$
$$\frac{1}{\epsilon_\perp} = \overline{\frac{1}{\epsilon(z)}}, \tag{12}$$

and the line over a function denotes an average over the period.

Equations (4) and (11) give

$$\epsilon_M(\theta,\omega) = \epsilon_\perp(\omega)\cos^2\theta + \epsilon_\parallel(\omega)\sin^2\theta. \tag{13}$$

The bulk energy loss function of the superlattice is then given by

$$L_b(\theta,\omega) = -\mathrm{Im}\,\frac{1}{\epsilon_M(\theta,\omega)}. \tag{14}$$

The dispersion relation for a long-wave bulk plasmon propagating in the θ direction then is

$$\frac{\epsilon_\perp(\omega)}{\epsilon_\parallel(\omega)} = -\tan^2\theta. \tag{15}$$

It can be easily shown that the macroscopic dielectric tensor

$$\hat{\epsilon} = \begin{pmatrix} \epsilon_\parallel & 0 & 0 \\ 0 & \epsilon_\parallel & 0 \\ 0 & 0 & \epsilon_\perp \end{pmatrix}, \tag{16}$$

which is defined by

$$\mathbf{D}_M = \hat{\epsilon}\,\mathbf{E}_M, \tag{17}$$

by virtue of Eq. (1) gives exactly Eq. (13). This justifies our notations of $\epsilon_\parallel$ and $\epsilon_\perp$.

III. SURFACE ENERGY LOSS FUNCTION OF SEMI-INFINITE SUPERLATTICE

A. Separation of the surface response

To separate the surface excitations of a semi-infinite superlattice from the bulk ones the following general procedure may be pursued. Let us define the surface response of a semi-infinite system as its response to the perturbation provided by the fluctuations of an external charge density located outside the system. Then, assuming that the system occupies the $z < 0$ half-space, we can write

$$\Delta\phi_{\mathrm{ext}}(\mathbf{r}) = 0,\ z < 0, \tag{18}$$

since there is no external charge inside the system. For definite $\mathbf{q}_\parallel$, Eq. (18) gives the only decreasing into the solid solution

$$\phi_{\mathrm{ext}}(\mathbf{r}) = Ae^{i\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel + q_\parallel z}. \tag{19}$$

But it is evident that the behavior of the external field in vacuum does not affect the semi-infinite system at all.[15] Then we may seek for the response of the system to the external perturbation (19) valid in the whole space notwithstanding the exponential increase of Eq. (19) into vacuum. The field induced by the system in vacuum satisfies the equation

$$\Delta\phi_{\mathrm{ind}}(\mathbf{r}) = 0,\ z > 0, \tag{20}$$

for there is no induced charge in vacuum. Then we can write for $z > 0$

$$\phi(\mathbf{r}) = \phi_{\mathrm{ext}}(\mathbf{r}) + \phi_{\mathrm{ind}}(\mathbf{r}) = (Ae^{q_\parallel z} + Be^{-q_\parallel z})e^{i\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}, \tag{21}$$

since the potential induced by the system must decrease into vacuum.

Equation (21) makes it clear that the surface response of a semi-infinite system is completely characterized by $B(\omega, q_\parallel)/A(\omega, q_\parallel)$ with A and B defined by Eq. (21).

In recent work[16] we have shown that the surface energy loss function of an arbitrary semi-infinite homogeneous in the XY plane system is given by

$$L_s(q_\parallel,\omega) = -\pi i \lim_{\substack{q_z\to +iq_\parallel \\ k_z\to -iq_\parallel}} (q_z - iq_\parallel)(\epsilon^{-1})_2(q_z,k_z,q_\parallel,\omega), \tag{22}$$

where

$$(\epsilon^{-1})_2(q_z,k_z,q_\parallel,\omega) = \frac{\epsilon^{-1}(q_z,k_z,q_\parallel,\omega) - \epsilon^{-1}(q_z,k_z,q_\parallel,-\omega)}{2i}$$

and $\epsilon^{-1}(q_z,k_z,q_\parallel,\omega)$ is defined by

$$\phi(q_z,q_\parallel,\omega) = \int \epsilon^{-1}(q_z,k_z,q_\parallel,\omega)\phi_{\mathrm{ext}}(k_z,q_\parallel,\omega)dk_z.$$

It follows from Eq. (22) that $L_s(q_\parallel,\omega)$ is given by the $\exp(-q_\parallel z)$ component of ϕ in response to the $\exp(q_\parallel z)$ component of ϕ_{ext}. Then we can write by use of Eqs. (21)

and (22)

$$L_s(q_\parallel,\omega) = -\frac{1}{2}\,\mathrm{Im}\,\frac{B(q_\parallel,\omega)}{A(q_\parallel,\omega)}. \tag{23}$$

We shall see in Sec. IV that for a half-space occupied by a homogenous medium with dielectric function $\epsilon(\omega)$, Eq. (23) gives $-\mathrm{Im}\ 1/[1+\epsilon(\omega)]$, in compliance with the conventional result.[9,17]

B. Macroscopic approach

First, let us consider the surface response of a semi-infinite superlattice macroscopically. Then we must find ϕ of the form (21) as a response to the external perturbation (19). We can write by use of Eqs. (1), (17), and (16)

$$\left[\epsilon_\perp \frac{d^2}{dz^2} - \epsilon_\parallel q_\parallel^2\right]\phi(z) = \left[\frac{d^2}{dz^2} - q_\parallel^2\right]\phi_{\mathrm{ext}}(z) = 0, \quad z<0, \tag{24}$$

where the explicit form (19) of ϕ_{ext} was used. Equations (24) and (21) give

$$\phi(z) = C\exp\left(\sqrt{\frac{\epsilon_\parallel}{\epsilon_\perp}}\,q_\parallel z\right), \qquad z<0,$$
$$\phi(z) = A\exp(q_\parallel z) + B\exp(-q_\parallel z),\ z>0, \tag{25}$$

where the square root must be taken with the positive real part.

The boundary conditions are

$$C = A+B,$$
$$\epsilon_\perp\sqrt{\frac{\epsilon_\parallel}{\epsilon_\perp}}\,C = A-B, \tag{26}$$

where the first is the continuity of the parallel component of the electric field and the second is the continuity of the normal component of the dielectric displacement. Equations (23) and (26) give for the surface energy loss function of a semi-infinite superlattice

$$L_s(\omega) = -\mathrm{Im}\,\frac{1}{1+\sqrt{\epsilon_\parallel(\omega)\epsilon_\perp(\omega)}}, \tag{27}$$

where the square root must be taken with the positive imaginary part, which is the consequence of the sign selection in Eq. (25).

The equation for the eigenfrequency of the surface plasmon is

$$\sqrt{\epsilon_\parallel(\omega)\epsilon_\perp(\omega)} = -1. \tag{28}$$

C. Microscopic approach

Expression (27) for the surface energy loss function of a semi-infinite superlattice was derived macroscopically without accounting for the local-field effects. Meanwhile the short-wave components of the response to the long-wave perturbation may, generally speaking, contribute to both the field equations and the boundary conditions. The object of the present section is to demonstrate that for the model under study the rigorous microscopic derivation does not change the final result (27).

Let us consider the response of our system to the external perturbation (19). After Fourier transform, we have

$$\phi(z) = \sum_G \phi_G \exp[i(G+q_z)z],\ z<0,$$
$$\phi(z) = A\exp(q_\parallel z) + B\exp(-q_\parallel z),\ z>0, \tag{29}$$

$$\sum_G [(G+q_z)(G'+q_z) + q_\parallel^2]\ \epsilon(G-G')\ \phi_{G'} = 0. \tag{30}$$

Let us regard Eq. (30) as a system of linear algebraic equations for the determination of ϕ_G. Then this system is compatible if we choose $q_z = q_0$ such that the determinant of this system is zero. After some algebraic transformations and using the properties of determinants, we can express all of ϕ_G by ϕ_0

$$\phi_G = \lim_{q_z\to q_0} \frac{\epsilon^{-1}_{G0}(q_z)}{\epsilon^{-1}_{00}(q_z)}\ \phi_0. \tag{31}$$

The boundary conditions are

$$\sum_G \phi_G = A+B,$$
$$\epsilon(z=0)\sum_G (G+q_z)\phi_G = q_\parallel(A-B). \tag{32}$$

By using Eqs. (12)

$$q_0/q_\parallel = \cot\theta_0 = -i\sqrt{\frac{\epsilon_\parallel}{\epsilon_\perp}}. \tag{33}$$

After substituting Eq. (31) into Eqs. (32), taking advantage of the equality

$$\lim_{q_z\to q_0}\frac{\epsilon^{-1}_{G0}(q_z)}{\epsilon^{-1}_{00}(q_z)} = -\frac{q\exp(i\theta_0)}{G}\left(\frac{1}{\epsilon}\right)(G) \times(\sin^2\theta_0\ \epsilon_\parallel + i\sin\theta_0\cos\theta_0\ \epsilon_\perp), \tag{34}$$

which follows from Eq. (11) and some algebraic transformations, we shall obtain the boundary conditions

$$\phi_0 = A+B,$$
$$\phi_0\ \epsilon_\perp\sqrt{\frac{\epsilon_\parallel}{\epsilon_\perp}} = A-B, \tag{35}$$

which are the same as Eq. (26) within notations. This proves that the expression (27) for the surface energy loss function is valid within the microscopical derivation.

Let us note that the agreement between the microscopic and macroscopic results are due to the fact that only the long-wave components of ϕ and $D_z/q_\parallel$ do not vanish in the long-wave limit, which can be seen from Eqs. (32) and (34). In contrast, all components of $E_z/q_\parallel$ are nonzero, but it does not affect the boundary conditions (35).

IV. COMPARISON WITH THE PREVIOUS THEORIES

In this work, we applied the method of inverting the microscopic dielectric matrix to find the macroscopic dielectric function and the excitation spectrum of a superlattice with an arbitrary varying dielectric function of the constituents. Of course, this technique is strictly equivalent to others (the transfer matrix method and the method of the direct solving of the Maxwell's equations) in both the quantum and the classical regimes. Specifically, it may be stated, for example, that the eigenvalues of the matrix (7) represent the excitation spectrum found in Ref. 4 by solving Maxwell's equations with boundary conditions at interfaces for *any* $\mathbf{q}$ as long as $\epsilon(G)$ is the Fourier transform of the step function taking two distinct values in the corresponding layers. In this section, we show this explicitly in the long-wave limit we are dealing with.

If the constituent dielectric function $\epsilon(z)$ is constant over the period, then by Eqs. (12) $\epsilon_{\|} = \epsilon_{\perp} = \epsilon$. Equation (13) then gives $\epsilon_M = \epsilon$. For the surface loss function we then have the well known result[9,17]

$$L_s(\omega) = -\mathrm{Im}\frac{1}{\epsilon(\omega)+1}. \tag{36}$$

The dispersion relation for the bulk plasmon in a superlattice constituted by alternating layers of two distinct dielectric functions ϵ_1 and ϵ_2 with thicknesses d_1 and d_2, reads[4]

$$(\epsilon_1^2+\epsilon_2^2)\sinh(q_{\|}d_1)\sinh(q_{\|}d_2) + 2\epsilon_1\epsilon_2\{\cosh(q_{\|}d_1)\cosh(q_{\|}d_2) - \cos[(d_1+d_2)q_z]\} = 0. \tag{37}$$

In the long-wave limit (37) gives

$$(\epsilon_1^2+\epsilon_2^2)d_1d_2 + \epsilon_1\epsilon_2[d_1^2+d_2^2+(d_1+d_2)^2\cot^2\theta] = 0. \tag{38}$$

In our approach, we have for this system

$$\epsilon_{\|} = \overline{\epsilon(z)} = \frac{1}{d_1+d_2}(d_1\epsilon_1+d_2\epsilon_2),$$
$$\frac{1}{\epsilon_{\perp}} = \overline{\frac{1}{\epsilon(z)}} = \frac{1}{d_1+d_2}\left(\frac{d_1}{\epsilon_1}+\frac{d_2}{\epsilon_2}\right). \tag{39}$$

Substituting Eq. (39) into Eq. (15) we obtain just Eq. (38).

The dispersion relation of the surface plasmon in this superlattice is[4]

$$\sinh(q_{\|}d_1)[\epsilon_2\cosh(q_{\|}d_2) - \sinh(q_{\|}d_2)]\epsilon_1^2 + (\epsilon_2^2-1)\cosh(q_{\|}d_1)\sinh(q_{\|}d_2)\epsilon_1 + \epsilon_2\sinh(q_{\|}d_1)[\epsilon_2\sinh(q_{\|}d_2) - \cosh(q_{\|}d_2)] = 0. \tag{40}$$

In the long-wave limit, Eq. (40) gives

$$d_1\epsilon_2\epsilon_1^2 + (\epsilon_2^2-1)\epsilon_1 d_2 - \epsilon_2 d_1 = 0. \tag{41}$$

Substituting Eq. (39) into Eq. (28), we obtain just Eq. (41).

V. DISCUSSION

Above, we have considered infinite and semi-infinite superlattices constituted by an arbitrary varying periodical dielectric function. In local theory, in the long-wave limit we have found the general solutions for the bulk and the surface dielectric response of these systems.

In the bulk case, Eq. (13) shows that the macroscopic dielectric function of a superlattice in the long-wave limit depends on the direction of the wave propagation. As a consequence, even in the long-wave limit, the bulk plasmon of a superlattice has a directional "spatial dispersion" (15). It must be noted that this spatial dispersion is obtained in a model, in which the intrinsic dielectric function of the constituents of the superlattice has no spatial dispersion. The directional spatial dispersion of the bulk plasmon in a superlattice is due, of course, to the anisotropy of this system.

Let us consider a metallic superlattice with the Drude-type constituent dielectric function

$$\epsilon(\omega,z) = 1 - \frac{\omega_p^2(z)}{\omega(\omega+i0_+)} = 1 - \frac{4\pi e^2 n(z)}{m\omega(\omega+i0_+)}, \tag{42}$$

where $n(z)$ is the electron density, $\omega_p(z)$ is the local plasma frequency, and m and e are the mass and the charge of the electron. We assume no intrinsic damping in $\epsilon(\omega,z)$. If we calculate $\epsilon_{\|}$ and $\epsilon_{\perp}$ by Eqs. (12), then $\epsilon_{\|}$ will not include the imaginary part, but for $\epsilon_{\perp}$ we have[18]

$$\begin{aligned}\mathrm{Im}\frac{1}{\epsilon_{\perp}(\omega)} &= \frac{\omega^2}{c}\mathrm{Im}\int_0^c \frac{dz}{\omega^2-\omega_p^2(z)+i0_+} \\ &= -\frac{\omega^2}{c}\int_0^c \delta[\omega^2-\omega_p^2(z)]dz \\ &= -\frac{\omega^2}{c}\sum_k \left|\frac{d\omega_p^2(z_k)}{dz}\right|^{-1},\end{aligned} \tag{43}$$

where z_k are the roots of the equation

$$\omega_p(z_k) = \omega \tag{44}$$

within a period.

It can be seen from Eq. (43) that for ω in the interval

$$\min \omega_p(z) < \omega < \max \omega_p(z) \quad (45)$$

the imaginary part of $\epsilon_\perp$ is nonzero. By using Eqs. (13), the macroscopic dielectric function of a superlattice will also have a nonzero imaginary part for all θ, except for $\theta = 90°$. The bulk [Eq. (14)] and the surface [Eq. (27)] loss functions will have the nonzero linewidths, which describe the damping of the bulk and the surface collective excitations in the superlattice. We want to point out once more that this result is inherent for superlattices with a continuously varying constituent dielectric function, for only in this case Eq. (44) has a solution(s). Let us note that this result is obtained without including the damping into the constituent dielectric function.

It is interesting to note that the neglect of the local-fields effects [using Eq. (5) instead of Eq. (4)], which is a good approximation for crystalline solids,[10] is thoroughly unacceptable for superlattices, because in this approximation the superlattice is replaced by a homogeneous medium with an average dielectric function. It may be said that all the effects in superlattices which distinguish them from the homogeneous media are the local-field effects.

It can be seen from Eq. (13) that if $\theta = 90°$, then the superlattice response is that of a homogeneous medium with dielectric function equal to an average over the period of the constituent dielectric function. So we have proved this result in the long-wave limit for quite an arbitrary constituent dielectric function. For superlattices with two distinct constituents it was shown in Ref. 19.

In Fig. 1, the real and the imaginary parts of the macroscopic dielectric function and the bulk energy loss function of a model metallic superlattice with the constituent dielectric function (42) are depicted.

The macroscopic dielectric function of the metallic superlattice with varying charge density shown in Fig. 1(a), is given by Eq. (13), where

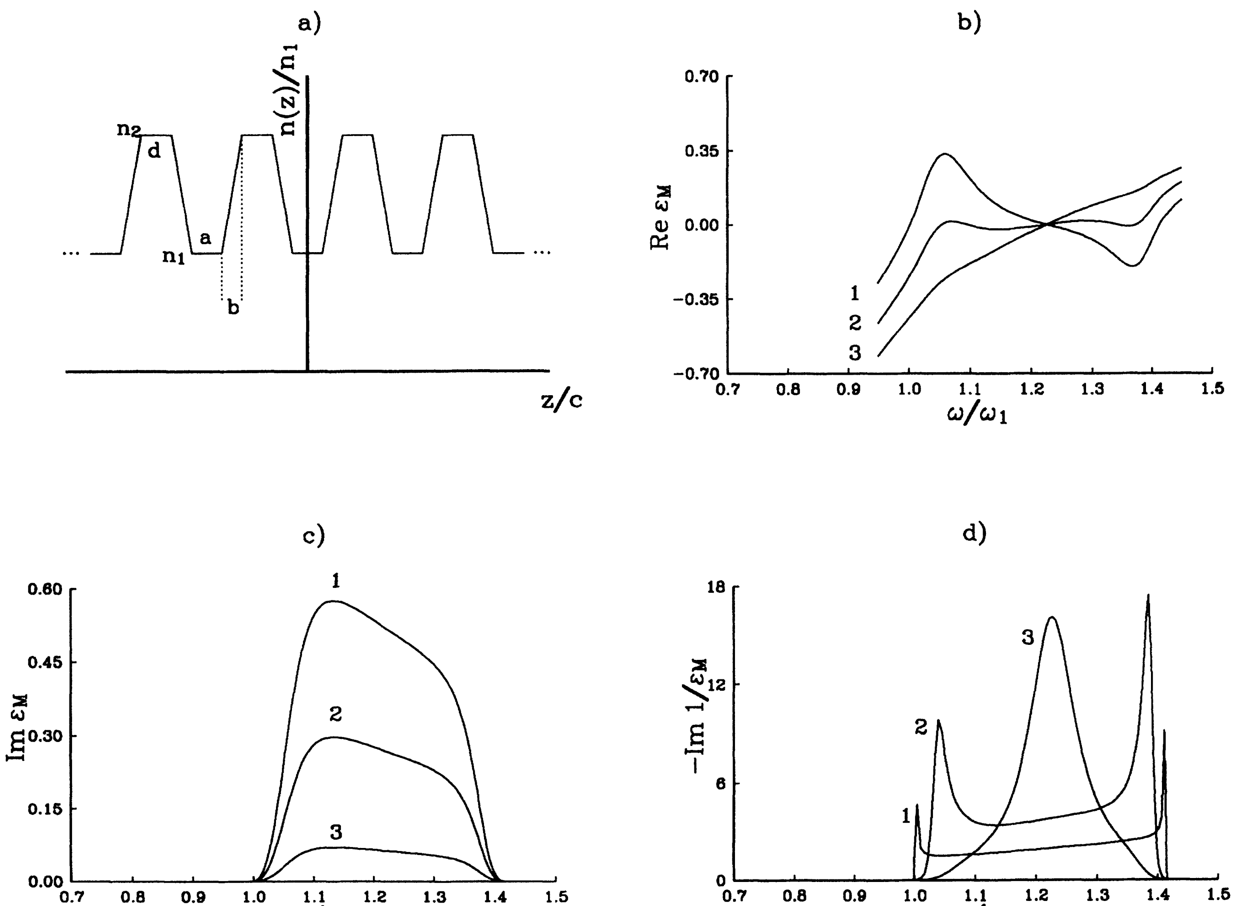

FIG. 1. Infinite superlattice. (a) The model electron density of a superlattice with the Drude-type constituent dielectric function Eq. (42). The parameter values are $a/c = d/c = 0.3$, $b/c = 0.2$, and $n_2/n_1 = 2$. The period of the superlattice is $c = a + 2b + d$. (b) and (c) are the real and the imaginary parts of the macroscopic dielectric functions of the superlattice, respectively. (d) The bulk energy loss functions of the superlattice. The labels 1, 2, and 3 denote the wave propagation with the angle to the superlattice axis $\theta = 10°$, $45°$, and $70°$, respectively. ω_i are the plasma frequencies corresponding to the electron densities n_i.

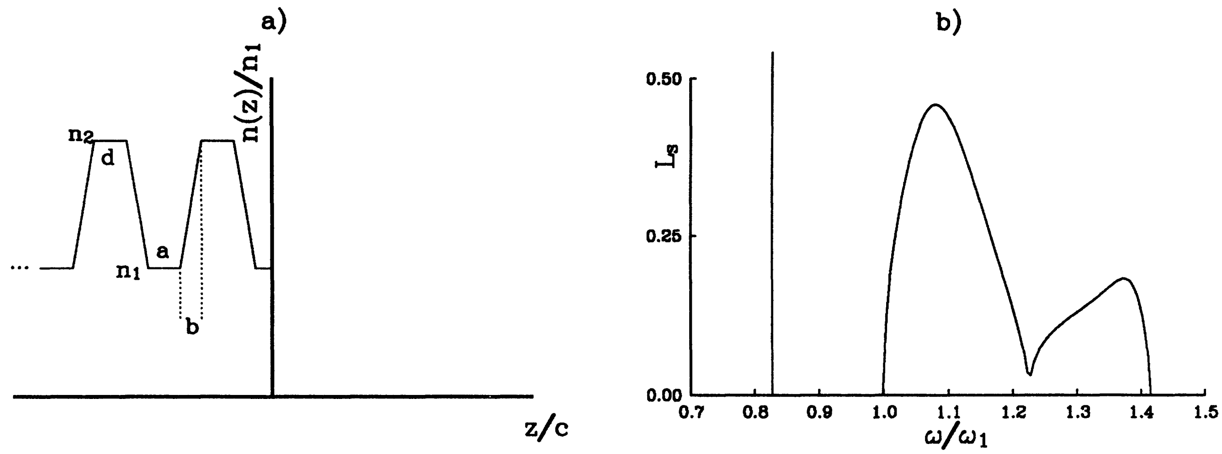

FIG. 2. Semi-infinite superlattice. (a) The model electron density of a superlattice with the Drude-type constituent dielectric function Eq. (42). The parameter values are the same as in Fig. 1. (b) The surface energy loss function obtained by Eq. (27) for the superlattice with the electron density plotted in Fig. 2(a). The vertical line shows the δ peak corresponding to the surface loss inherited from the loss spectrum of the semi-infinite homogeneous medium (see discussion in the text).

$$\frac{1}{\epsilon_\perp(\omega)} = \frac{\omega^2}{c}\left[\frac{-2b}{\omega_2^2-\omega_1^2}\left(\ln\left|\frac{\omega_2^2-\omega^2}{\omega^2-\omega_1^2}\right| -i\pi\left[\begin{matrix}1 & \omega\in(\omega_1,\omega_2)\\ 0 & \omega\notin(\omega_1,\omega_2)\end{matrix}\right]\right)+\frac{a}{\omega^2-\omega_1^2}+\frac{d}{\omega^2-\omega_2^2}\right],$$

$$\epsilon_\parallel(\omega) = 1-\frac{\omega_{\rm av}^2}{\omega^2}, \tag{46}$$

$$\omega_{\rm av}^2 = \frac{1}{c}[a\omega_1^2 + d\omega_2^2 + b(\omega_1^2+\omega_2^2)].$$

In Fig. 2(b), we present the surface loss function (27) of the metallic semi-infinite superlattice, the charge density of which is plotted in Fig. 2(a). The constituent dielectric function is chosen the same as in the bulk case [Eq. (42)]. In the surface case we observe the same effect of the finite damping of peaks in the loss function without the intrinsic damping of the constituent dielectric function, as in the bulk one. The vertical line in Fig. 2 (b) shows the δ-peak corresponding to the surface loss inherited from the loss spectrum of the semi-infinite homogeneous medium, which can be demonstrated by tending the charge density in the period to a constant. The energy position of this peak then tends to $\omega_p/\sqrt{2}$, as it must be.[20] In our example this peak is not broadened, since its energy does not satisfy Eq. (45). If we take the damping constant in the Drude dielectric function of the constituents of the superlattice nonzero, this peak will have a finite width. Two other peaks in Fig. 2(b) are inherent to the superlattice with continuously varying dielectric function and they disappear in the homogeneous case or their widths tend to zero in the case of a superlattice with distinct constituents.

VI. CONCLUSION

We have considered the superlattices constituted by a variable dielectric function $\epsilon(\omega,z)$, which is an arbitrary periodic function of coordinate z. In the long-wave limit, using the local theory, we have obtained the analytical solutions for the dielectric response and the excitation spectrum of this system in the infinite and semi-infinite cases.

Both the bulk and the surface response of this system are characterized by the two bulk quantities: the average over the period of the functions $\epsilon(\omega,z)$ and $1/\epsilon(\omega,z)$.

We have shown that in the long-wave limit the bulk plasmon of the superlattice has a directional spatial dispersion, which is the consequence of the anisotropy of the system.

The specific damping of the surface and the bulk collective modes in the superlattices with continuously varying constituent dielectric functions, which is not present in superlattices with distinct layers, is obtained and discussed.

It is shown that the local-field effects represent all the properties, which distinguish the superlattices from the homogeneous media.

Our theory is also applicable to superlattices with distinct constituents. In this case, it gives the results that are in accordance with those known from the literature.

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