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# Nonlocal extensions of the electromagnetic response of plasmonic and metamaterial structures

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Nonlocal effects, requiring wave-vector- ($q$-) dependent dielectric response functions, are becoming increasingly important in studies of plasmonic and metamaterial structures. The phenomenological hydrodynamic approximation is the simplest and most often used model but with limited applicability to problems involving surface plasmons. We show here that the $d$-function formalism, exact to first order in $q$, is a powerful and simple-to-use alternative, which allows for exact nonlocal extensions of local calculation schemes, e.g., finite-difference time-domain methods, without code changes. It is also extendable to order $q^2$, and we demonstrate this by comparing with various earlier *ab initio* calculations and experiments as well as by performing our own random-phase-approximation calculations (valid for all $q$) of the surface-plasmon dispersions for simple metals with various electron-gas densities. Finally we show that this hydrodynamic-extended $d$-function formalism can also be applied to arbitrary plasmonic/metamaterial structures as long as the nonflat interfaces can be modeled as effective media films.

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## I. INTRODUCTION

In many studies of plasmonic and/or metamaterial systems, their electromagnetic behavior can be obtained from a local dielectric response model, which assumes that the excitations involve a vanishingly small momentum exchange, i.e., a vanishing wave vector [1]. Although this is usually an excellent approach, commonly used in optical studies of materials and employed in various simulation codes [including finite-difference time-domain (FDTD) methods], in some cases it fails. For instance, the well-known local Drude formula for the dielectric function of simple metals [1,2] $\varepsilon(\omega) = 1 - \omega_p^2/\omega^2$, where $\omega_p$ is the plasma frequency, is an excellent approximation at high (e.g., visible) frequencies. However, it completely fails at very low frequencies (far IR and below) where the nonlocality of the response becomes explicit [3], i.e., $\varepsilon(q,0) \approx 1 + q_{TF}^2/q^2$. This immediately leads to the well-known, static Thomas-Fermi screening, and its positivity also leads to BCS superconductivity in metals [4], for instance. Clearly the nonlocality completely controls the physics in this case.

In this paper we first discuss two simple approximate schemes for nonlocal extensions of the local Drude model: the well-known phenomenological hydrodynamic approximation (HDA) and the *ab initio* $d$-function formalism of Feibelman, exact to first order in $q$. We show how the $d$-function formalism can be used to generalize any local calculation scheme (e.g., FDTD) by mapping the nonlocal effects onto an effective coating, having a complex dielectric function. Thus, any local calculations or simulations that include this film become automatically nonlocal to the same order. We also show how the $d$-function formalism can be extended to order $q^2$ (in the spirit of the HDA) and demonstrate the validity of this extension by comparing to *ab initio* calculations and experimental data of the surface-plasmon (SP) dispersion on simple metallic surfaces as reported in the literature. Additionally, we compare to our own *ab initio* random-phase-approximation (RPA) calculations (valid for all $q$) of the surface-plasmon dispersions in thick metallic slabs for various electron-gas densities. Finally, we demonstrate that the HDA can be used to estimate the parameters of our $d$-function formalism extension and show that this HDA-extended $d$-function formalism can also be applied to arbitrary plasmonic/metamaterial structures as long as the nonflat interfaces can be modeled as effective media films.

## II. SIMPLE NONLOCAL EXTENSIONS

The simplest nonlocal model is the HDA, which extends the local Drude model as follows [5,6]:

$$\varepsilon(q,\omega) = \varepsilon_b - \frac{\omega_p^2}{\omega(\omega + i\gamma) - \beta q^2}, \tag{1}$$

with $\beta = \frac{3}{5}v_F^2$, where $v_F$ is the Fermi velocity, $\varepsilon_b$ is the bound electron contribution, and $\gamma$ is the loss parameter (rate of electron scattering with phonons and defects). This equation reduces both to the Drude model for high frequency as well as to the Thomas-Fermi model for vanishing frequency, showing that the Thomas-Fermi wave vector is given by $q_{TF} = \omega_p/\sqrt{\beta}$.

Despite many successful applications, the HDA has been known to be inadequate at describing the dispersion of SPs [7,8]. An alternative successful approach has been proposed by Feibelman, who in his seminal papers of the late 1970s not only pioneered *ab initio* computer simulations of the electromagnetic effects of metallic interfaces, but also developed a simple nonlocal parametrization of the surface problem with his *ab initio* $d$-function formalism [9,10]. He showed that the nonlocal electromagnetic response of a simple (alkali-) metal surface is fully described (to first order in $q$) by the standard Fresnel formulas with a small adjustment involving a complex surface response function called the $d$ function [11], given by $d(\omega) = \int dz\, z\, \delta\rho(z,q=0,\omega)/\int dz\, \delta\rho(z,q=0,\omega)$. Here, $\delta\rho$ is the in-plane charge density

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induced by a long-wavelength field at frequency $\omega$ with $z$ perpendicular to the surface—thus, the $d$ function physically represents the centroid of the induced charge density, which, for alkali metals, is located outside of the surface. We explicitly write the formula for the modified reflection coefficient of Feibelman in the following section for a particular geometry but first report a salient result.

Using this formalism, a very simple nonlocal expression for the SP dispersion of metals can be obtained by requiring divergence of the reflection coefficient [12], which yields the well-known result [10],

$$\omega(q) = \omega_{sp}\left[1 - \tfrac{1}{2}d(\omega)q\right], \tag{2}$$

where $\omega_{sp} = \omega_p/\sqrt{\varepsilon_b + \varepsilon_0}$. Since $d(\omega) > 0$ for all alkali metals [8], (2) demonstrates that the initial slope of the dispersion for small $q$ is negative for these metals. It is important to note that the $d$ functions are obtained using nonretarded formalisms without any loss of accuracy and have been calculated for various simple (alkali) and complex (noble) metals [7,13–16]. The $d$-function formalism has thoroughly been confirmed by electron energy loss spectroscopy experiments [17,18] and provides deep insight into the SP dynamics at small $q$. Thus, any first-order calculation of nonlocal surface physics should incorporate the $d$ function.

## III. GEOMETRIC MAPPING

We first consider the lowest-order extension of the nonlocal response of a metallic surface using the $d$ function. Since the $d$ function is a surface response function, the nonlocal physics that it encapsulates may be conveniently mapped onto the local surface dielectric function of a thin fictitious film (of thickness $\Delta d$) placed on top of this metal surface. Then conventional Fresnel optics can be used to calculate the reflection coefficient for this system, whose geometry consists of an outside dielectric [$\varepsilon_0$], a surface layer [$\varepsilon_s(\omega)$], and a metal [$\varepsilon(\omega)$] in the configuration depicted in Fig. 1.

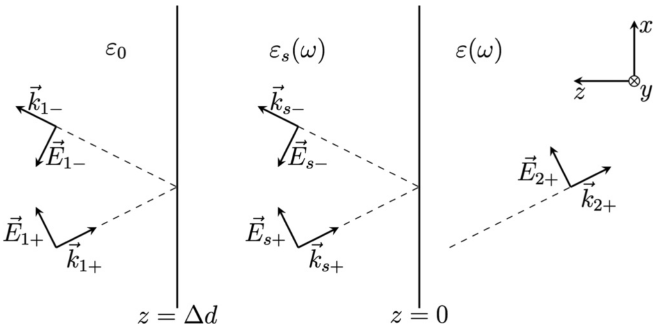

FIG. 1. Reflection geometry for $p$-polarized monochromatic light incident on a layered system from the positive $z$ direction. A slab of thickness $\Delta d$ and permittivity $\varepsilon_s(\omega)$ acts as an intermediate layer (index $s$), confined by a dielectric half-space with permittivity $\varepsilon_0$ (index 1) and a metallic half-space with Drude permittivity $\varepsilon(\omega)$ (index 2). The plane of incidence is the $xz$ plane.

We derive $r_p$ for a monochromatic transverse wave ($p$ polarized) incident upon the system described above. The electric field in the three regions can be written as

$$\vec{E}_1(\vec{r},t) = \vec{E}_{1+}e^{i(\omega t - qx + k_1 z)} + \vec{E}_{1-}e^{i(\omega t - qx - k_1 z)}, \tag{3}$$

$$\vec{E}_s(\vec{r},t) = \vec{E}_{s+}e^{i(\omega t - qx + k_s z)} + \vec{E}_{s-}e^{i(\omega t - qx - k_s z)}, \tag{4}$$

$$\vec{E}_2(\vec{r},t) = \vec{E}_{2+}e^{i(\omega t - qx + k_2 z)}, \tag{5}$$

where $\vec{E}_{j\pm}$ is the electric-field vector of the incoming (+) or outgoing (−) wave in the outside dielectric ($j = 1$), surface layer ($j = s$), or metal ($j = 2$). The respective wave vectors are $\vec{k}_{j\pm} = (q, \mp k_j)$, where $k_j = \sqrt{(\omega/c)^2\varepsilon_j - q^2}$ is the $z$ component and $q$ is the in-plane component. Using the standard boundary conditions across an interface ($D_z$ and $E_x$ continuous), the reflection coefficient is

$$r_p \approx \frac{\varepsilon(\omega) - \varepsilon_0\frac{k_2}{k_1} + i\frac{\varepsilon(\omega)\varepsilon_0}{\varepsilon_s(\omega)k_1}q^2\Delta d - i\alpha_-(\omega)\Delta d}{\varepsilon(\omega) + \varepsilon_0\frac{k_2}{k_1} - i\frac{\varepsilon(\omega)\varepsilon_0}{\varepsilon_s(\omega)k_1}q^2\Delta d + i\alpha_+(\omega)\Delta d}, \tag{6}$$

where $\alpha_\pm(\omega) = [\frac{\varepsilon(\omega)\varepsilon_0}{k_1}(\frac{\omega}{c})^2 \pm \varepsilon_s(\omega)k_2]$ and an overall phase has been neglected.

Now, the nonlocal reflection coefficient for this geometry but without the presence of a film was shown by Feibelman [10] to be

$$r_p = \frac{\varepsilon(\omega) - \varepsilon_0\frac{k_2}{k_1} + i\frac{[\varepsilon(\omega)-\varepsilon_0]}{k_1}q^2 d(\omega)}{\varepsilon(\omega) + \varepsilon_0\frac{k_2}{k_1} - i\frac{[\varepsilon(\omega)-\varepsilon_0]}{k_1}q^2 d(\omega)}, \tag{7}$$

where positive $d(\omega)$ lies above the metal surface and, as in (6), index 1 corresponds to the outer dielectric and index 2 corresponds to the metal. In order to identify the mapping we require that (6) is identical to (7), which, with the natural assumption that $\varepsilon_s(\omega) \sim \Delta d$ and for sufficiently small $\Delta d$, requires

$$\varepsilon_s(\omega) = \frac{\varepsilon(\omega)\varepsilon_0\Delta d}{[\varepsilon(\omega) - \varepsilon_0]d(\omega)}. \tag{8}$$

Thus, one has a simple prescription for extending any local calculation or simulation, such as FDTD, into the small-$q$ nonlocal domain: Before performing a simulation, one must first add a fictitious dielectric film of finite thickness $\Delta d$ to any metallic surface of the structure. This film must have the complex frequency-dependent (but local and nonretarded) dielectric function $\varepsilon_s(\omega)$ given by (8). The simulations now include nonlocal effects, accurate to lowest order in $q$.

The HDA version of this mapping of nonlocal effects into structural extensions (local films) was first proposed and studied in Ref. [6] and recently in Ref. [19]. This paper provides justification and extension of these studies based on the $d$-function formalism. In fact, (8) reduces to the corresponding equation in Ref. [19] after a formula for the $d$ function, appropriate for the HDA, is used [6,10]

$$d_{HDA}(\omega) = \frac{i}{q_{pl}} = \sqrt{\beta/[\omega_p^2/\varepsilon_b - \omega(\omega + i\gamma)]}, \tag{9}$$

where $q_{pl}$ is the plasmon normal wave vector, obtained from the condition $\varepsilon(q,\omega) = 0$ with $\varepsilon(q,\omega)$ given by (1).

## IV. HDA-EXTENDED *d*-FUNCTION FORMALISM

A higher-order nonlocal expression for the SP dispersion of metals, approximately valid for all $q$, can be obtained by quadratically extending (2), which yields

$$\omega(q) = \omega_{sp}\left[1 - \tfrac{1}{2}d(\omega)q + bq^2\right]. \tag{10}$$

This ansatz follows from the Taylor expansion of $\omega(q)$ with the next term expected to be of $O(q^2)$. We refer to this as the hydrodynamic term because of how it has been defined in the literature, e.g., in (1). Ultimately, we show that the coefficient $b$ has the same physical characteristics as the relevant term in the HDA.

Figure 2 shows that one can nearly perfectly fit *ab initio* calculations, such as the local density approximation (LDA) [17], using (10) with $b$ as a free parameter. In obtaining the fit represented by the red-dotted line in Fig. 2, we calculated Re[$\omega$] in (10) using the complex $d$ function for potassium as given in Ref. [7]. Our fit is very good for the monopole SP mode (lower branch). In addition, it qualitatively describes the multipole SP mode (upper branch), which is related to a pole structure of the $d$ function [1,7]. Due to finite losses incorporated in the $d$ function, the two branches of the SP modes are connected in a characteristic $s$-shaped way, similar to how the two monopole SP branches are connected in the retarded limit [1]. The fit represented in Fig. 2 by the red-solid line employs the same procedure as for the red-dotted line but models the pole structure in the $d$ function with a Lorentzian form with zero damping, thus, capturing the entire behavior of the undamped multipole branch, starting from $q = 0$. Note, that the experimental and simulation data for the upper branch for small $q$ are closer to the red-solid line, which shows that the $d$ function overestimates losses for this branch when using our HDA-extended $d$-function formalism. The SP dispersion for $b = 0$ also is shown in Fig. 2 for comparison (black dashed-dotted lines). Finally, the pure HDA SP dispersion, given by (10) with a vanishing $d$ function and $b = \beta/2\omega_{sp}^2$, is shown as a blue-dotted line. Note that the HDA alone fails to describe the SP dispersion, in particular, its negative slope for small $q$.

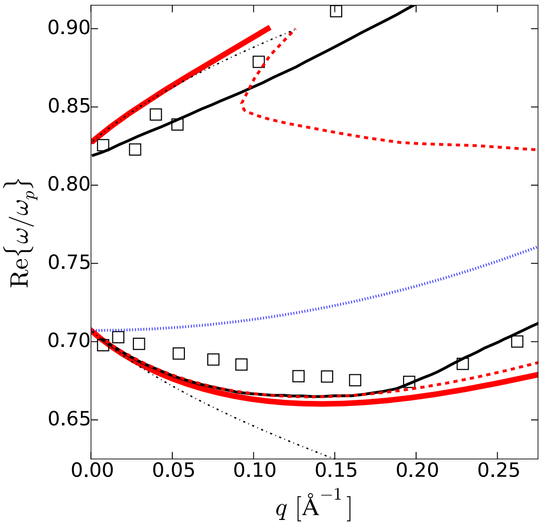

FIG. 2. Surface-plasmon dispersion for potassium: experimental results (open squares, from Ref. [17]); LDA calculation (black-solid lines, from Ref. [17]); calculation employing the extended model (10) in conjunction with the full complex $d$ function (red-dotted line); calculation employing the extended model in conjunction with the *lossless* *d* function (red-solid lines). The black dashed-dotted lines represent the dispersion for $b = 0$. The pure HDA result is shown as a blue-dotted line. Data for the $d$ function are obtained from Ref. [7].

Equation (10), our HDA-extended $d$-function formalism, is the main result of this paper. In the next section, we provide evidence that $b$ has scaling properties identical to the appropriate hydrodynamic term by studying its behavior as a function of the electron-gas density.

## V. FULL *AB INITIO* NONLOCAL RESPONSE OF METALLIC SLABS

The nonlocal effects for arbitrary $q$ can be calculated exactly in the RPA for varying electron density. Specifically, we have performed RPA calculations based on the formalism of Ref. [20] applied to a metallic slab of finite thickness [17,21], which employs the known self-consistently calculated ground states (for various electron densities) of Lang and Kohn [22]. Then the electron-density $\delta\rho(z;\omega)$, induced by an external perturbation $V_{\text{ext}}$, is calculated using the following integral equation:

$$\delta\rho(z;\omega) = \int dz'\,\chi_0(z,z';\omega) \times\left[V_{\text{ext}} + \int dz''\,V_c(z',z'')\delta\rho(z'';\omega)\right], \tag{11}$$

where $\chi_0(z,z';\omega)$ is the single electron susceptibility and $V_c(z,z') = e^2/|z - z'|$. We have solved (11) for four different electron densities, parametrized by Wigner-Seitz radii $r_s = 2, 3, 4, 5$. Subsequently, the induced density is used to calculate the complex reflection coefficient via $r(q,\omega) = \int dz\, e^{qz}\delta\rho(z;\omega)$.

Figure 3 shows color maps of log[Im$\{r(q,\omega)\}$] plotted vs frequency $\omega$ and wave-vector $q$, each for different electron densities. Maxima of Im$\{r(q,\omega)\}$ occur at the surface-plasmon condition, and those maxima are marked as open circles on these maps. Due to the finite thickness of the slab and the resulting coupling between SPs on opposite sides, there is a well-known split of the dispersion at small momenta (here for $q < 0.05$ Å$^{-1}$) with the upper and lower branches representing the antisymmetric and symmetric coupled SP modes, respectively. The maxima corresponding to these split modes have not been marked but are visible due to color coding.

For larger $q$ (of interest here) a single branch exists, representing the SP mode of a single surface. The thin-solid lines represent the results of RPA calculations for semi-infinite metallic systems, available in the literature [17,18], calculated for similar electron densities. There is an excellent agreement between our slab and the semi-infinite calculations for $q > 0.1$ Å$^{-1}$ [23], demonstrating the usefulness of the slab scheme. Additionally, in Fig. 3 we also show the measured dispersion relations for surfaces of bulk alkali metals with similar densities when available [17,18]. These are in qualitative agreement with our calculations in the $q > 0.1$ Å$^{-1}$ range, in

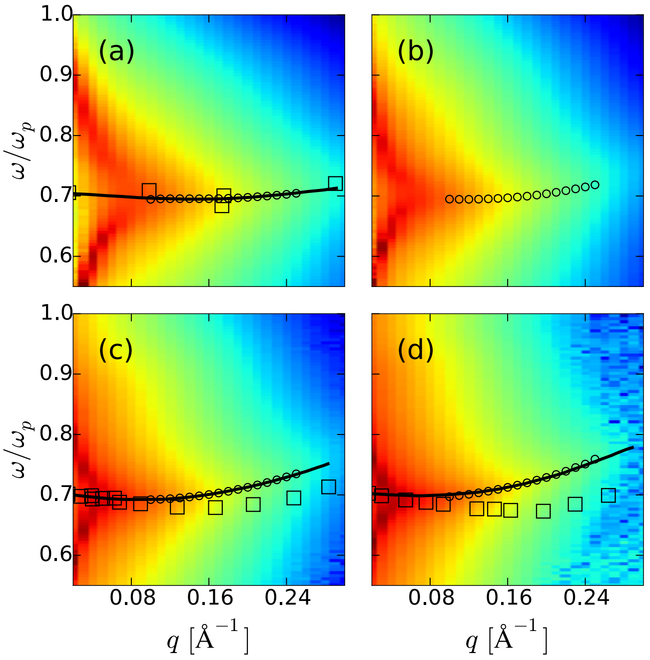

FIG. 3. Color maps of $\log[\mathrm{Im}\{r(q,\omega)\}]$ from RPA calculations of metallic jellium slabs for (a) $r_s = 2$, (b) $r_s = 3$, (c) $r_s = 4$, and (d) $r_s = 5$. The circles represent maxima of $r(q,\omega)$, which occur at the SP dispersion condition. The solid lines represent the RPA calculated results for semi-infinite jellium [17], and squares are the experimental results [17] for (a) $r_s = 2.07$, (c) $r_s = 3.93$, and (d) $r_s = 4.86$. No simulation or experimental data are available in literature for $r_s = 3$.

particular, at higher electron densities. Note, that no simulation or experimental data exist in the literature for $r_s = 3$.

To investigate the density scaling properties in detail, we normalize and parametrize the SP relations as suggested by (10),

$$\frac{\omega(q)}{\omega_{sp}} = 1 - aq + bq^2. \tag{12}$$

This equation was employed to fit the calculated dispersions of Fig. 3 using the least-squares method. Figure 4(a) shows that this fit is excellent, confirming that our HDA-extended $d$-function formalism is well motivated.

Figure 4(b) shows the fit-extracted parameters $a$ (black triangles) and $b$ (red squares) plotted as a function of $r_s$. Clearly, $b$ scales linearly with $r_s$ with the red-dashed line as a guide to the eye. This linear scaling is consistent with the behavior of $\beta$ in the HDA, implying that the HDA is a good model for $b$. This can be shown by substituting (1) into the local version of the reflection coefficient (7), searching for the vanishing denominator of $r_p$, and matching the result with the functional form of (12). This procedure yields

$$b = \beta/2\omega_{sp}^2 \propto r_s. \tag{13}$$

Similarly, assuming the HDA form of the $d$ function (9) and performing the same analysis, we obtain

$$a = \sqrt{\beta}/2\omega_{sp} \propto \sqrt{r_s}. \tag{14}$$

The black-dashed line represents the scaling given by (14) and shows that the HDA scaling again agrees well with the fit obtained from (12) (black-solid triangles). This demonstrates that the HDA can be used to estimate the electron-density scaling of the $a$ and $b$ parameters of our HDA-extended $d$-function formalism.

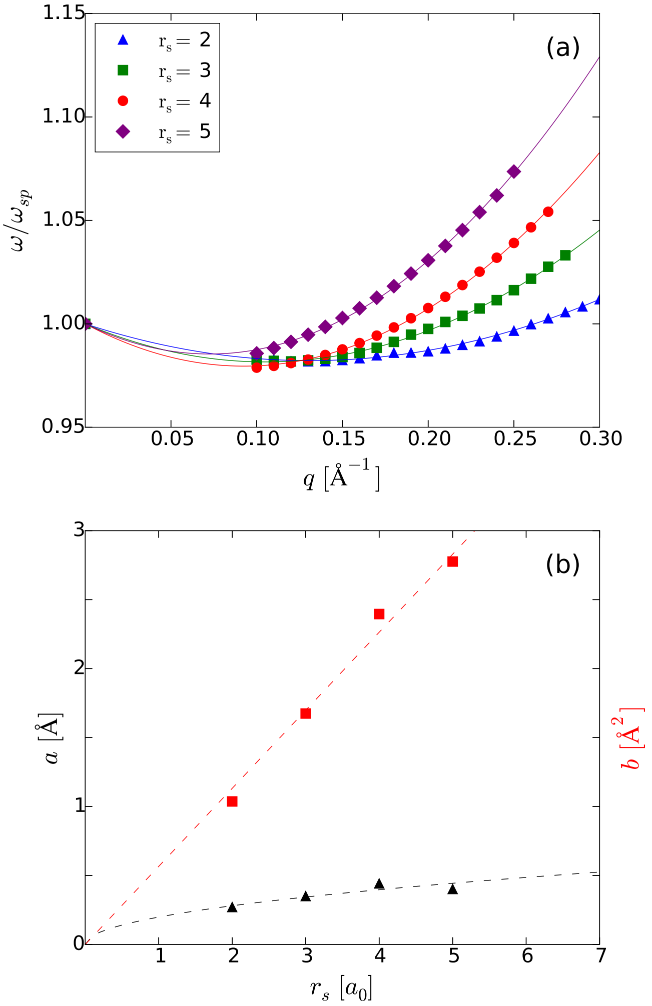

FIG. 4. (a) SP dispersions for metallic jellium slabs with $r_s = 2,3,4,5$: RPA calculations (symbols), least-squares fits to these calculations using (12) (solid lines). (b) $a$ vs $r_s$: RPA fits (black triangles) and HDA model fit to points (black-dashed line); $b$ vs $r_s$: RPA fits (red squares) and HDA model fit to points (red-dashed line).

Relations (13) and (14) above also suggest that, for systems with the effective electron mass changed ($m = \eta m_0$) rather than the electron density, the following scaling holds:

$$b \propto 1/\eta, \tag{15}$$

and

$$a \propto 1/\sqrt{\eta}. \tag{16}$$

## VI. HDA-EXTENDED *d*-FUNCTION FORMALISM FOR METAMATERIALS

The analysis presented in Sec. V was for metallic systems with flat interfaces. However, the simple scalings developed there are expected to hold for structures with nonflat interfaces, treated as flat effective media films. This idea is supported by Pendry *et al.* [24] in their important paper, which pioneered the metamaterial concept. They showed that the effective dielectric function of a three-dimensional cubic metallic wire array in the long-wavelength limit is given by the Drude form but with the plasma frequency renormalized by the effectively reduced density (electrons are confined to wires and thus occupy a smaller volume of the array; $r_s$ scaling) and by an increased effective mass (an inductive effect; $\eta$ scaling). As a result of both renormalizations, the wire medium was shown to have its plasma frequency dramatically shifted down from the UV to the radio range. The same phenomenon is expected for other metamaterial structures, e.g., for those with nonflat surface morphology.

In this context, one can rewrite the formula (7) into the conventional local form [25]

$$r_p = \frac{\varepsilon(q,\omega) - \varepsilon_0}{\varepsilon(q,\omega) + \varepsilon_0}, \tag{17}$$

but require that the SP dispersion relation is still given by (12). This leads to an effective dielectric function of the material, which now has the characteristic modified-Drude form

$$\varepsilon(q,\omega) = \varepsilon_b - \frac{\omega_p^2}{\omega(\omega + i\gamma) + \Sigma}, \tag{18}$$

where

$$\Sigma = 2\omega_{sp}^2(aq - bq^2), \tag{19}$$

with $a$ and $b$ defined as in (12) and which can be estimated by the HDA.

Thus, accounting for nonlocal effects amounts to the following very simple transformation in the effective dielectric function formula:

$$\omega(\omega + i\gamma) \rightarrow \omega(\omega + i\gamma) + \Sigma. \tag{20}$$

The same transformation can be used in the more general Drude-Lorentz form of the effective dielectric function,

$$\varepsilon(\omega) = \varepsilon_b + \sum_m \frac{\omega_{pm}^2}{\omega_{0m}^2 - \omega(\omega + i\gamma)}. \tag{21}$$

Thus, the full procedure to implement the nonlocal extensions is as follows. First, a local simulation of a metamaterial structure should be performed, for example, by employing FDTD. Second, the effective local dielectric function should be extracted, for example, by using the standard procedure developed in Ref. [26], and fitted with the Drude-Lorentz form (21). Finally, the transformation (20) provides the desired extension. A similar procedure was applied in the recent study of the electron scattering in the presence of phonons and plasmons, which requires nonlocal corrections [27], except in this case a simple linear model of $\Sigma$ was used instead of (19), and a different method was employed to extract the dielectric function.

## VII. CONCLUSIONS

In this paper, we first developed a mapping of the *ab initio* Feibelman theory ($d$-function formalism) of nonlocal corrections onto a thin film, having a local and complex dielectric function. The resulting calculated response is exact to lowest order in $q$, and any local calculation, including FDTD, can be applied to such a modified structure, without any need for code changes. Next, we proposed a simple large-$q$ extension of the $d$-function formalism in the spirit of the HDA and demonstrated the usefulness of this scheme in describing SP dispersions for very large $q$. We supported this HDA-extended $d$-function formalism with specific *ab initio* RPA calculations for metallic films with flat surfaces for various electron densities. We have also shown that the HDA, even though itself incapable of describing SP dispersions, can be used to estimate the parameters of our extension. Finally, we have generalized the HDA-extended $d$-function formalism to arbitrary plasmonic/metamaterial structures in which nonflat interfaces can be modeled as effective media films.

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