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Physical nature of volume plasmon polaritons in hyperbolic metamaterials

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Abstract: We investigate electromagnetic wave propagation in multilayered metal-dielectric hyperbolic metamaterials (HMMs). We demonstrate that high- k propagating waves in HMMs are volume plasmon polaritons. The volume plasmon polariton band is formed by coupling of short-range surface plasmon polariton excitations in the individual metal layers.

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1. Introduction

Hyperbolic metamaterials (HMMs) are subwavelength structures such as rod arrays [1–4] or multilayers [5,6] made to imitate "indefinite media," a special case of extreme anisotropy where diagonal elements of the permittivity tensor have different signs (e.g., $\epsilon_x = \epsilon_y < 0$ and $\epsilon_z > 0$) [7]. Such media are unusual because their dispersion relation $\omega^2/c^2 = (k_x^2 + k_y^2)/\epsilon_z + k_z^2/\epsilon_{x,y}$ is hyperbolic rather than elliptical [Fig. 1(a)]. In the idealization that this relation holds for all $k_{x,y,z}$, different signs of $\epsilon_{x,y}$ and ϵ_z result in propagating solutions for waves with infinitely large wave vectors, i.e., $k^2 \gg \epsilon_{x,y,z}\omega^2/c^2$. Such waves (the "high- k modes" [5–9]) are evanescent in isotropic media but become *propagating* in indefinite media. As a result, the photonic density of states (DOS) becomes unbounded, giving rise to new physical phenomena such as broadband spontaneous emission enhancement [5, 6, 10], anomalously high heat-transfer capabilities [11], and optical simulation of metric signature transitions [12]. Practical applications, e.g., near-field [13–15] and far-field subwavelength imaging (hyperlensing) [8, 16] and highly absorptive "darker than black" coatings that benefit from surface roughness [17], were also proposed.

However, attributing the physics of HMMs to high- k modes raises a few concerns. First, even though structural elements of an HMM, e.g., layer thicknesses in Fig. 1(b), may be much smaller than the vacuum wavelength at the frequency of interest, the wavelength of high- k modes, $2\pi/k$, will eventually become even smaller. Therefore any real structure will have a

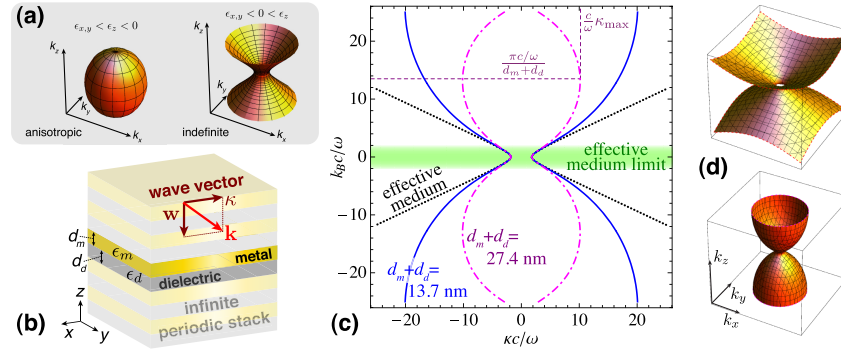


Fig. 1. (a) Isofrequency surfaces in the dispersion relation $(k_x^2 + k_y^2)/\epsilon_z + k_z^2/\epsilon_{x,y} = \omega^2/c^2$ for conventional anisotropic medium ($\epsilon_{x,y,z} > 0$) and indefinite medium ($\epsilon_{x,y} < 0$ and $\epsilon_z > 0$). (b) An infinite multilayer HMM with wave vector decomposition used in the paper (which is schematic in that w and k can be complex). (c) Comparison between the actual multilayer dispersion relation [Eq. (4)] for $d_m + d_d = 13.7$ and 27.4 nm with the effective-medium dispersion relation for $\rho = 0.17$. The materials have $\epsilon_m = -17.2$ (gold [18]) and $\epsilon_d = 2.59$ (alumina [19]) for $\lambda = 715$ nm [5]. (d) Isofrequency surfaces for the actual dispersion relation (*bottom*) vs. effective medium approximation (*top*).

range of k where it is no longer “subwavelength” and can no longer be described by the effective medium approach [2,5,6], displaying pronounced optical nonlocality effects [20,21]. Hence, all conclusions about unbounded DOS in real HMMs become unreliable, and significant deviations from effective-medium predictions are expected in real HMMs [21–24], even when optical nonlocalities are taken into account in the homogenization theory [25].

Furthermore, the effective index approach does not explain the transition from a conventional material on a smaller (microscopic) scale to an HMM on a larger scale, and fails to provide any insight into the physical nature of high- k modes other than to assert that they must be bulk propagating waves. Such insight would be crucial to explore the extent to which these modes can be used as information carriers. Since the structures involved are metal-dielectric, it is strongly believed that high- k modes are plasmonic in nature. So various groups call them *multilayer plasmons* [26], *Bloch plasmon polaritons* [27], or *volume plasmon polaritons* (VPPs) [28], while other groups relate them to leaky waves in anisotropic waveguides [13,29,30] or to surface polaritons [4,31]. In metal-dielectric multilayers, it is stipulated that these VPPs arise from surface plasmon polaritons (SPPs) at layer interfaces [14,32]. However, the direct connection between an SPP at a single interface and a VPP in the multilayer HMM is still to be drawn, and the prevailing coupling mechanism between the SPPs is still not quite clarified [33].

In this paper, we provide a direct proof that a band of bulk propagating waves (VPPs) with large wave vectors in periodic multilayer HMMs originates from coupling of SPPs in the individual metal layers by virtue of the Bloch theorem. When the reflection coefficient of metal layers is replaced by a simple pole expansion that only preserves short-range SPP (SRSPP) excitations, correct dispersion relation of an HMM is recovered throughout its entire 1D Brillouin zone. We conclude that “high- k waves” in HMMs can be physically understood as “Bloch VPPs”, and can be seen as a distinct type of plasmonic excitations.

The paper is organized as follows. In Section 2, we review the basics of wave propagation in metal-dielectric multilayers, and discuss the dispersion relation for bulk waves in multilayer HMMs. In Section 3, we employ the pole expansion formalism to preserve only the SRSPPs in the metal layers, and show that bulk VPPs originate from surface-bound SRSPPs in the metal layers via the Bloch theorem. Finally, in Section 4 we summarize.

2. High- k bulk propagating waves in HMMs

We consider an infinite periodic metal-dielectric HMM in Fig. 1(b) where metal layers with permittivity ϵ_m and thickness d_m alternate with dielectric layers with permittivity ϵ_d and thickness d_d . The dispersion relation can be determined by the standard transfer matrix approach [34]. The transfer matrix for one period of the structure can be written as

$$M_1 = \frac{1}{T_m} \begin{bmatrix} T_m^2 - R_m^2 & R_m \\ -R_m & 1 \end{bmatrix} \begin{bmatrix} e^{iw_d d_d} & 0 \\ 0 & e^{-iw_d d_d} \end{bmatrix}. \quad (1)$$

The reflection and transmission coefficients of a metal layer are given by the Airy formulas,

$$R_m = r_{dm} + t_{dm} r_{md} t_{md} e^{2iw_m d_m} [1 - r_{md}^2 e^{2iw_m d_m}]^{-1}, \quad T_m = t_{dm} t_{md} e^{iw_m d_m} [1 - r_{md}^2 e^{2iw_m d_m}]^{-1}. \quad (2)$$

In Eqs. (1)–(2), $w_j = [\epsilon_j \omega^2 / c^2 - \kappa^2]^{1/2}$ ($j = m, d$) is the z -directed component of the wave vector in metal or dielectric. (To fix the definition, we take $\text{Im } w_j \geq 0$; if $\text{Im } w_j = 0$ we take $\text{Re } w_j \geq 0$.) The reflection and transmission coefficients r_{ij} and t_{ij} at the $(i|j)$ interface are given by standard Fresnel formulas for p -polarized light,

$$r_{md} = \frac{w_m \epsilon_d - w_d \epsilon_m}{w_m \epsilon_d + w_d \epsilon_m}, \quad r_{dm} = \frac{w_d \epsilon_m - w_m \epsilon_d}{w_d \epsilon_m + w_m \epsilon_d}, \quad t_{md} = \frac{2w_m \sqrt{\epsilon_m \epsilon_d}}{w_m \epsilon_d + w_d \epsilon_m}, \quad t_{dm} = \frac{2w_d \sqrt{\epsilon_d \epsilon_m}}{w_d \epsilon_m + w_m \epsilon_d}. \quad (3)$$

Assuming for now that the metal is lossless, we can conveniently label the plane wave components by a real tangential component κ lying in the x – y plane, as it does not vary across layers. Since the multilayer is periodic in z , it supports propagating Bloch waves with the wave vector $k^2 = k_B^2 + \kappa^2$. The relation between k_B and κ is given by the Bloch theorem [25, 32, 33]:

$$\frac{\text{Tr } M_1}{2} = \cos[k_B(d_m + d_d)] = \cos(w_m d_m) \cos(w_d d_d) - [(\eta + \eta^{-1})/2] \sin(w_m d_m) \sin(w_d d_d), \quad (4)$$

where $\eta = (\epsilon_m w_d) / (\epsilon_d w_m)$. As shown earlier (see, e.g., [26]), expanding Eq. (4) up to $(w_j d_j)^2$ reduces it to the hyperbolic dispersion equation, $\omega^2 / c^2 = k_B^2 / \epsilon_{x,y} + \kappa^2 / \epsilon_z$ where $\epsilon_{x,y} = \rho \epsilon_m + (1 - \rho) \epsilon_d < 0$ and $\epsilon_z^{-1} = \rho \epsilon_m^{-1} + (1 - \rho) \epsilon_d^{-1} > 0$ are the effective permittivity tensor components, and $\rho \equiv d_m / (d_m + d_d)$ is the metal filling fraction.

Figure 1(c) compares this approximate dispersion relation with the exact result of Eq. (4). As expected [25, 26], we see that VPPs in multilayer HMMs are indeed Bloch waves in a periodic metal-dielectric multilayer, approaching a hyperbolic dispersion relation for smaller κ but deviating from it as κ increases. The cut-off in the range of possible κ , $\kappa_{\max} \propto 1 / (d_m + d_d)$ [24], puts a limit on the photonic DOS in real multilayers [see Fig. 1(d)]. For $\kappa \lesssim \kappa_{\max}$, where the VPPs reach wavelengths as small as the layer thicknesses, the multilayer behaves like a photonic crystal rather than like a homogeneous effective medium, displaying a band structure. The hyperbolic dispersion relation comes from the local curvature of the propagation band near the Γ -point of the 1D Brillouin zone. In the HMM regime, the high- k waves are backward-propagating [15, 21, 30].

The condition where the effective-medium approximation is valid, $w_j d_j \ll 1$, can be regarded as a special case of a “subwavelength condition” for HMMs, implying that neither the phase nor the amplitude of a propagating wave should vary significantly across the thickness of any layer in the structure. Unlike conventional media where the subwavelength condition can simply be $d_j / \lambda \ll 1$ due to the elliptical dispersion relation restricting the range of possible κ , the “HMM subwavelength condition” breaks down for some large κ no matter how thin the layers are.

3. Pole expansion for surface plasmon polaritons in metal layers

Bloch waves in crystals are typically formed from elementary excitations in single elements (“atoms”) that couple in periodic systems to form bands. We can thus understand the underlying physics of HMMs by identifying the nature of the elementary excitations behind VPPs.

Although it has been generally understood that such excitations in metal-dielectric multilayers must be plasmonic in nature [21, 26, 27], a direct connection between SPPs at individual metal-dielectric interfaces and VPPs in HMMs can be established. To do so, we replace physical metal layers in an HMM by "fictitious" layers that only support one plasmonic mode, and compare the wave propagation properties in this fictitious structure with those in the original HMM.

From Eq. (1) we see that the Fresnel coefficients R_m and T_m play a central role in the dispersion relation, so the excitations we seek to characterize must be identified in these coefficients. Poles in the Fresnel coefficients signal the presence of guided wave-type excitations in multilayers. For example, SPPs at a single metal-dielectric interface are seen as poles in r_{dm} and t_{dm} [see Eq. (3)], but their wave number κ_{md} , as given by the expression $w_m(\kappa_{md})\epsilon_d + w_d(\kappa_{md})\epsilon_m = 0$, is on the order of $(\omega/c)\sqrt{\epsilon_d}$ at optical frequencies. What we must clarify is how the wave vectors of VPPs in HMMs become enormously large ($\kappa \gg \kappa_{md}$).

In thin metal layers, comprising two metal-dielectric interfaces, SPPs are known to couple either symmetrically or antisymmetrically, producing long-range SPPs (LRSPPs) or short-range SPPs (SRSPPs), respectively. Both are described by the pole condition in Eqs. (2),

$$1 - r_{md}^2 e^{2i w_m d_m} = 0 \quad (5)$$

Asymmetric coupling in SRSPPs makes their wave vectors larger than for LRSPPs. Assuming that $\kappa^2 \gg \epsilon_{m,d} \omega^2 / c^2$, we solve Eq. (5) to obtain a limit for the SRSPP wave number for $d_m \rightarrow 0$: $\kappa_{sp} \approx (\ln r)/d_m$, $r = (\epsilon_m - \epsilon_d)/(\epsilon_m + \epsilon_d)$. We see that κ_{sp} becomes very large as d_m decreases, confirming that the involvement of SRSPP in the VPP formation is a well-justified conjecture.

To confirm the validity of this conjecture and to see that SRSPPs are crucial in the dispersion relation of HMMs, we replace the expressions for T_m and R_m in Eq. (1) by their pole expansion around the SRSPP pole at κ_{sp} : $T_m = \tau_m/(\kappa - \kappa_{sp})$, $\tau_m \approx (r^{-1} - r)/(2d_m)$. Note that as $\kappa \rightarrow 0$, $T_m \rightarrow -\tau_m/\kappa_{sp} \approx 1$, in agreement with the complete transmission of a very thin ($d_m \ll c\sqrt{\epsilon_m}/\omega$) metal film. The corresponding pole expansion for R_m would give $-\tau_m/(\kappa - \kappa_{sp})$. However, to stay consistent with vanishing reflectivity for a very thin metal film at normal incidence we add a constant to this expression

$$R_m = -\tau_m/(\kappa - \kappa_{sp}) - (\tau_m/\kappa_{sp}), \quad T_m^2 - R_m^2 = -2\tau_m^2/[\kappa_{sp}(\kappa - \kappa_{sp})] - (\tau_m/\kappa_{sp})^2. \quad (6)$$

Substituting Eq. (6) in Eq. (1), instead of Eq. (4) we get

$$\cos[k_B(d_m + d_d)] \approx [1 - (\kappa - \kappa_{sp})/(2\tau_m)] e^{i w_d d_d} + [(\kappa - \kappa_{sp})/(2\tau_m)] e^{-i w_d d_d}. \quad (7)$$

In Fig. 2(a), it can be seen that Eq. (7) provides a good approximation for Eq. (4). The agreement remains reasonable for ρ up to 0.5, getting slightly better for smaller ρ , and persists for lossy metals. Interestingly, omitting the second terms in Eqs. (6) [using just $R_m = -\tau_m/(\kappa - \kappa_{sp})$] can also achieve a decent agreement between Eqs. (7) and (4) in the range of larger $c\kappa/\omega$ with κ near κ_{sp} , which is the region of most VPPs [Fig. 2(b)]. However, adding the correction term drastically improves the agreement near $k_B = 0$ where $c\kappa/\omega$ is smaller, making the pole approximation accurate throughout the entire Brillouin zone.

It is also remarkable that the pole expansion (6) continues to give a correct dispersion relation for smaller $\rho < \epsilon_d/(\epsilon_m + \epsilon_d)$, where $0 < \epsilon_{x,y} < \epsilon_z$, and the structure is no longer an HMM but an extremely anisotropic medium. As seen in Fig. 2(c), the exact dispersion relation has two branches [21]. Only the inner, low- k , forward-wave branch is recovered by the effective medium approximation, which may lead to an erroneous conclusion that high- k waves disappear for smaller filling fractions. In fact, high- k waves are still contained in the other (outer) branch, which gets pushed to higher κ , outside of the validity of the condition $w_m d_m \ll 1$. This backward-wave branch is correctly recovered by the pole expansion, indicating (in line

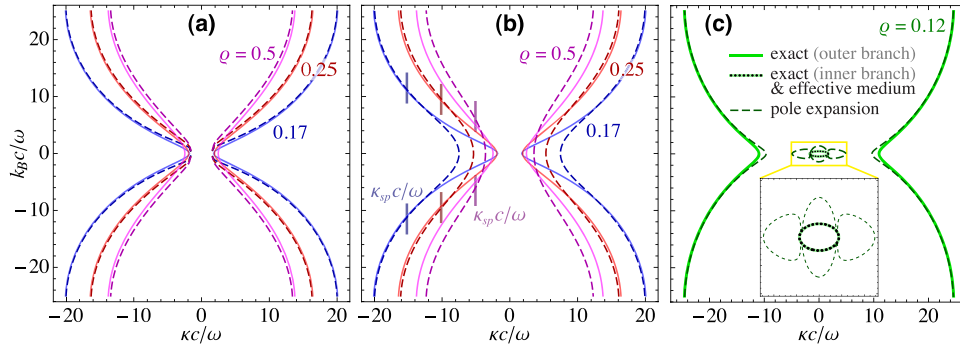


Fig. 2. Comparison between Eq. (4) (*solid*) and the coupled-SRSPP approximation given by Eq. (7) (*dashed*) for $d_m + d_d = 13.7$ nm and $p = 0.17, 0.25$, and 0.5 for (a) the pole expansion as in Eq. (6) and (b) the pole expansion without the added constant terms (the vertical marks showing the location of $\kappa_{sp}c/\omega$ for the respective filling fractions); (c) same as (a) for $p = 0.12$ (the effective medium approximation is indistinguishable from the inner branch of the exact relation). The materials are the same as in Fig. 1(c).

with the results of [21]) that VPPs in metal-dielectric multilayers also exist when the HMM requirements are not met.

For higher metal filling fractions, $p > 0.5$, it would be expected that the dispersion relation should be more accurately recovered by the pole expansion for a thin metal-dielectric-metal gap waveguide rather than a metal layer [33]. The corresponding expressions can be obtained from Eqs. (6)–(7) with the subscripts m and d interchanged. However, the agreement with the exact Eq. (4) is only slightly better than Eq. (7) for $0.5 < p < -\epsilon_m/(\epsilon_m + \epsilon_d) \approx 0.8$, so Eq. (7) remains a good approximation throughout the HMM parameter range.

Finally, note that there is surely more to the Fresnel coefficients R_m and T_m than SRSPPs. For example, LRSPPs are signaled by the presence of poles in those coefficients as well. However, the width of the LRSPP poles is much narrower in κ , and they do not have any influence on the dispersion relations at large $c\kappa/\omega$.

4. Conclusions

We have investigated electromagnetic wave propagation in multilayered metal-dielectric hyperbolic metamaterials (HMMs). Starting with the assumption that the only excitation supported by a thin metal film is an SRSPP, and coupling these excitations together through dielectric layers, we have reproduced the correct dispersion relation of “high- k waves” in a multilayer HMM throughout its 1D Brillouin zone. Hence, propagating waves in metal-dielectric multilayers, including HMMs, can indeed be understood as bulk plasmonic waves or VPPs arising from the coupling between SRSPPs in the constituent metal layers. In analogy with 2D arrays of plasmonic nanoparticles, where localized particle plasmons couple to form surface waves called Bloch SPPs [35], we can understand high- k propagating waves in HMMs as Bloch VPPs.

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