On the limits of the effective description of hyperbolic materials in the presence of surface waves

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Abstract

Here, we address the question of the validity of an effective description for hyperbolic metamaterials in the near-field region. We show that the presence of localized modes such as surface waves drastically limits the validity of the effective description, and requires revisiting the concept of homogenization in the near-field. We demonstrate, from exact scattering matrix calculations for multilayer hyperbolic structures, that one can find surface modes in spectral regions where the effective approach predicts hyperbolic modes only. Hence, the presence of surface modes which are not accounted for in the effective description can lead to physical misinterpretations in the description of hyperbolic materials and their related properties. In particular, we discuss in detail how the choice of the topmost layer affects the validity of the effective medium approach for calculating the local density of states and the super-Planckian thermal radiation.

Keywords: multilayer structure, hyperbolic material, effective medium theory, surface phonon polariton modes, super-Planckian radiation

With today’s nanotechnology it is possible to manufacture artificial composite materials at tiny scales which manifest optical properties that we do not encounter in nature. These media, also called metamaterials, are usually structured at the length scale, or below the wavelength, of photons. These media have been greatly exploited during recent decades to overthrow long standing paradigms in physics. For instance, new phenomena such as negative refraction [1], super-resolution [2] or reversed Doppler effects [3] were predicted for metamaterials.

One class of metamaterials, the so called indefinite or hyperbolic materials (HMs), has recently attracted much attention. This kind of material can not only support quasi-monochromatic surface modes [4–6], but can also have broadband hyperbolic modes. In fact, the dispersion relation of electromagnetic waves in these media can be represented by hyperbolic isofrequency curves, meaning that they support propagating modes having extremely large wavevectors far beyond the light line [7, 8]. Because of this property HMs are viewed as promising candidates to develop or improve breakthrough technologies, among which are sub-diffractive imagery (hyperlensing) [9–11], thermal management at nanoscale [12–14], near-field energy conversion [15], enhanced light emission [16], and quantum information systems [17]. The most simple realization of an HM is a plasmonic dielectric multilayer structure, but of course there are also other possible realizations of an HM (see [18, 19] for instance).

The exotic properties of HMs in the near-field regime are conventionally investigated within the framework of the
effective medium theory (EMT). Naturally, the question arises regarding under which conditions the EMT remains valid in the close vicinity of a composite material. In general, it is believed that for propagating modes the EMT is valid as long as the wavelength is much larger than the size of the unit-cell of the composite material. In the evanescent regime this condition imposes that the wavevector of the evanescent field has to be smaller than the inverse of the unit-cell size as well [20]. In the context of the spontaneous emission it has been shown that the EMT can greatly underestimate or overestimate [21–23] the Purcell factor when this condition is not met. In addition, when the composite materials support surface waves, one encounters a nonlocality which cannot be adequately described by the EMT [20, 24]. Another nonlocal effect can arise from the nonlocal response of the composite materials for very large wavevectors [25]. Therefore, a nonlocal effective description is needed to take such effects into account [26–30].

In this paper, we show that the presence of surface waves at the interface of the basic components of phonon-polaritonic multilayer HMs also requires revisiting of the concept of homogenization which otherwise, in the EMT framework, can lead to quantitatively incorrect and physically misinterpreted results if one estimates the hyperbolic bands from EMT [31, 32]. By comparing the exact calculation of the local density of states (LDOS) of the electromagnetic field at arbitrary distances above a multilayer HM with the predictions given by the EMT, we highlight the regions where the effective theory fails to describe properly the field outside the multilayer HM. In particular, we demonstrate from exact calculations the existence of surface modes inside the hyperbolic bands which are not predicted at all by the EMT. Hence, some properties attributed to the hyperbolicity of the material could be misinterpreted, or even worse could not exist any more. For example, large changes in the Purcell factors, thermal emission and Casimir–Lifshitz forces within the hyperbolic frequency bands could be due to such surface modes originating from the topmost layer, which are not taken into account in the EMT. Hence, the EMT would in this case give results which are quantitatively and qualitatively incorrect. We illustrate this fact by comparing the LDOS above a multilayer hyperbolic phonon-polaritonic structure with the heat transfer coefficient describing the radiative heat flux between two such structures.

To estimate the range of validity for the EMT in the presence of surface modes, we consider a 1D periodic structure composed of two materials, with one which naturally supports surface waves in the spectral range of interest. This structure is depicted in figure 1(a). One basis material is silicon carbide (SiC) and the other one is germanium (Ge). The polar medium SiC supports surface phonon polariton (SPP) modes in the mid-infrared at $\lambda_{SPP} = 10.3\ \mu m$ and its permittivity is given by

$$\varepsilon_{SiC} = \varepsilon_{\infty} \left( \frac{\omega_p^2 - \omega^2 - i\gamma\omega}{\omega_p^2 - \omega^2 - i\gamma\omega} \right)$$

following the Drude Lorenz model [33] with $\varepsilon_{\infty} = 6.7$, $\omega_p = 182.7 \times 10^{12}\ \text{rad}\ \text{s}^{-1}$, $\omega_T = 149.5 \times 10^{12}\ \text{rad}\ \text{s}^{-1}$ and $\gamma = 0.9 \times 10^{12}\ \text{rad}\ \text{s}^{-1}$. The dielectric permittivity of Ge in the same frequency range is constant and equal to $\varepsilon_{Ge} = 16$. According to the EMT, for $a < \lambda$, $2\pi c / \omega$ the equivalent homogenized medium to this structure is an uniaxial anisotropic medium with an effective permittivity parallel and perpendicular to the optical axis given by the expressions [34]

$$\varepsilon_{\parallel} = f\varepsilon_{SiC} + (1-f)\varepsilon_{Ge}$$

and

$$\varepsilon_{\perp} = \frac{f\varepsilon_{Ge} - (1-f)\varepsilon_{SiC}}{f\varepsilon_{Ge} + (1-f)\varepsilon_{SiC}}$$

with the volume filling fraction $f = l_1 / a$. The reflection coefficients for s- and p-polarized waves in the effective description are thus

$$r_s = \frac{k_0 - k_s}{k_0 + k_s} \quad \text{and} \quad r_p = \frac{\varepsilon_{\parallel} k_0 - k_p}{\varepsilon_{\parallel} k_0 + k_p}$$

with $k_0 = \sqrt{\omega^2 / c^2 - k_s^2}$, $k_s = \sqrt{\omega^2 / c^2 \varepsilon_{\parallel} - k^2}$ and $k_p = \sqrt{\omega^2 / c^2 \varepsilon_{\perp} - \varepsilon_{\parallel} k^2}$ the normal components of the wavevector in vacuum and in the medium for s and p waves. Here the wavevector $k$ is perpendicular to the optical axis. Therefore p-polarized electromagnetic waves (the so called extra-ordinary waves) in such an effective uniaxial medium fulfill the dispersion relation [35]

$$\frac{k^2}{\varepsilon_{\parallel}} + \frac{k^2}{\varepsilon_{\perp}} = \frac{\omega^2}{c^2}$$

If Re($\varepsilon_{\parallel}$) and Re($\varepsilon_{\perp}$) are both positive, equation (4) describes an elliptic dispersion curve in the $(k, k_z)$ plane, while in contrast if the effective permittivities have opposite signs, i.e., if Re($\varepsilon_{\parallel}$)Re($\varepsilon_{\perp}$) < 0, the dispersion curve described by equation (4) becomes hyperbolic. In the spectral range where Re($\varepsilon_{\parallel}$)Re($\varepsilon_{\perp}$) < 0 waves are propagating and are called hyperbolic modes. In figure 1(b) Re($\varepsilon_{\parallel}$) and Re($\varepsilon_{\perp}$) are plotted versus the frequency $\omega$ and the hyperbolic regions are highlighted in grey. For the chosen filling fraction $f = 0.4$ the first band $\Delta_1$ represents the case where Re($\varepsilon_{\parallel}$) < 0 and Re($\varepsilon_{\perp}$) > 0 whereas in the second band $\Delta_2$ the effective permittivities fulfill Re($\varepsilon_{\parallel}$) > 0 and Re($\varepsilon_{\perp}$) < 0.

To check the pertinence of the EMT predictions we use the exact S-matrix method [35–37] with a finite but large number of periods $N/2$. Beside, for our calculations we...
choose $N = 100$ layers where the last Ge-layer is assumed to be infinitely large extending to $z \to \infty$. For $N \to \infty$ it is well known that propagating waves in a periodic structure are Bloch waves and satisfy the Bloch mode dispersion relation [35]

$$\cos(k_z a) = -\frac{1}{2} \left( \frac{P_{22}k_{21}}{P_{11}k_{22} + P_{12}k_{12}} \right) \sin(k_z l_1) \sin(k_z l_2) + \cos(k_z l_1) \cos(k_z l_2),$$

with $P_{11} = P_{22} = 1$, $P_{12} = \epsilon_{\text{SiC}}$ and $P_{21} = \epsilon_{\text{Ge}}$.

To analyse the dispersion relation for the exact and the effective case we have plotted the isofrequency curves in the $\kappa$–$k_z$ plane from equations (4) and (5) for different frequencies in figure 2. Here we used the Bloch equation (5) which gives the exact isofrequency curves for the limiting case $N \to \infty$. For the frequencies $\omega = 1.6 \times 10^{14}$ rad s$^{-1}$ and $\omega = 1.9 \times 10^{14}$ rad s$^{-1}$ outside the hyperbolic regions the effective permittivities are both positive as shown in figure 1(b), and accordingly we get elliptical isofrequency curves. The difference between the principal axes of the black and orange curves stems from the fact that $\text{Re}(\epsilon_{\perp}) < \text{Re}(\epsilon_{\parallel})$ for the frequency range between $\Delta_1$ and $\Delta_2$, whereas for frequencies larger (smaller) than $\Delta_2$ ($\Delta_1$) $\text{Re}(\epsilon_{\parallel}) > \text{Re}(\epsilon_{\perp})$. The Bloch dispersion curves (solid line) for these frequency regions fit excellently the effective data (dashed line).

The green and blue curves in figure 2 are plotted for frequencies which are located in the hyperbolic bands. The dashed lines for $\omega = 1.55 \times 10^{14}$ rad s$^{-1}$ (blue) and $\omega = 1.7 \times 10^{14}$ rad s$^{-1}$ (green) illustrate the hyperbolic effective results where the blue curve is an example for a dispersion curve from the $\Delta_1$ band and the green one represents a dispersion curve from the $\Delta_2$ band. Here the Bloch dispersion coincides with the EMT result for small $\kappa$. For increasing values of $\kappa$ the solid curves show the existence of a $\omega$-dependent vertical asymptote as a consequence of Bloch equation (5), while the effective dashed curves do not have a maximal $\kappa$ value. This is a major difference between the behaviour of the structure predicted by the effective theory and by the rigorous theory.

Although the isofrequency curves deduced from the effective medium theory and from the Bloch theory coincide very well for $\kappa < 1/a$, as is well known, the exact calculations can deviate very strongly from the effective and Bloch results due to the fact that the structure can support surface modes which are not taken into account either in the Bloch theory or in the EMT. To demonstrate this fact we have plotted in figure 3 both the exact and the effective reflection coefficients (RCs) in the $\omega$–$\kappa$ plane in regions of small and large $\kappa$ values. In figures 3(a) and (c), where the exact RC is plotted, the black solid lines represent the boundaries of the Bloch bands given by equation (5) for propagating modes in the medium. Here the considered frequency range is still in the first Bloch band. Inside these Bloch areas the RC has large values and with increasing number $N$ these areas are filled due to an increasing number of discrete Bloch modes. The maximal $\kappa$ value for the contributing Bloch modes is therefore given by equation (5).

The RC for the EMT is plotted in figures 3(b) and (d). The black dashed lines mark the hyperbolic bands $\Delta_1$ and $\Delta_2$ where $\text{Re}(\epsilon_{\parallel})\text{Re}(\epsilon_{\perp}) < 0$. By comparing figure 3(a) with 3(b) with $\kappa_{\text{max}} = 10/a$ it can be seen that the EMT fails to reproduce the exact result for large $\kappa$ values, as already mentioned before. The boundaries for the Bloch bands change drastically whereas the size of the hyperbolic bands does not change with increasing $\kappa$ values. In figures 3(c) and (d) the exact and effective RC is plotted up to $\kappa_{\text{max}} = 1/a$. Here the RC calculated with the EMT nearly coincides with the exact result. The exact calculation shows discrete Bloch modes in the hyperbolic regime. This comes from the limited number of layers ($N = 100$). For $N \to \infty$ the discrete Bloch modes will fill the whole hyperbolic regime.

However, outside the Bloch areas in figure 3(a) two prominent features around $\omega_1 = 1.8 \times 10^{14}$ rad s$^{-1}$ (upper branch) and $\omega_2 = 1.6 \times 10^{14}$ rad s$^{-1}$ (lower branch) are visible with very high values for the RC. These modes are coupled
SPP modes stemming from the first SiC-layer on the top of the structure. These surface modes are damped on a scale much smaller than the period $a$ so that they do not ‘feel’ the periodic structure. The upper branch around $\omega_1$ has a very high RC and moreover high values up to very large $\kappa$. The SPP contribution becomes, therefore, relevant for very small distances $z$ to the periodic structure. In the exact calculations, as it can be seen in figure 3(a), the upper SPP branch is located in the second hyperbolic band $\Delta_2$ where the effective results do not show any SPP mode in the hyperbolic regimes. The contribution of this SPP mode around $\omega_1$ is still visible for $\kappa < 1/a$ as it can be seen in figure 3(c). The contribution of the coupled surface modes reduces but does not vanish. This qualitative difference between both approaches can lead to misinterpretations for very small distances to the structure.

In figures 4(a)–(d) we show the same results as in figures 3(a)–(d) but with Ge as topmost layer. Obviously, the surface mode contribution of the SiC-layer is shielded by the Ge-layer, resulting in a suppression of the surface mode contribution seen in figure 3. Hence, by choosing a ‘passive’ topmost layer (‘passive’ in the sense that the material does not support surface modes) the unwanted surface mode contribution in the hyperbolic band can be diminished. Furthermore, the qualitative validity of the EMT within the hyperbolic bands is in this case relatively good even for distances $z < a$, whereas the quantitative deviation from the exact results becomes important for distances on the order of $a$.

To estimate the range of validity of the EMT for a multilayer hyperbolic structure and to evaluate the disturbing
role played by the SPP mode, we have finally calculated the LDOS of the electromagnetic field for all filling fractions at different distances \( z \) above the SiC/Ge structure, both for the homogenized structure and for the real one. The results are plotted in figure 4. For \( z = 500 \) nm in figures 5(e) and (f), for both plots high LDOS values appear for large \( f \) at the lower right boundary of \( \Delta_2 \). This is due to surface mode resonances outside the hyperbolic bands where \( \text{Re}(\epsilon_\perp) \text{Re}(\epsilon_\parallel) = 1 \) and \( \text{Re}(\epsilon_\parallel) < 0 \). These conditions together with equation (2) lead to \( f > 0.5 \) as a necessary condition for having surface mode resonances outside the hyperbolic band within the EMT. Note that this surface mode contribution outside the hyperbolic band is well described by the EMT for \( z = 200 \) and 500 nm. The contribution of the coupled SPP mode of the first layer within the hyperbolic band \( \Delta_2 \) is very low and the effective calculations fit very well the exact results. However, when decreasing the distance \( z \) this contribution of the coupled SPP mode becomes larger, and eventually for \( z = 50 \) nm the effective LDOS is not able to mimic the exact LDOS as illustrated in figures 5(a) and (b). Here the contribution of the coupled SPP mode of the first layer is much larger than the contribution of the Bloch waves. Hence, from the EMT calculations one would expect a large LDOS stemming from the hyperbolic modes, which would be a wrong conclusion, since as we see from the exact results the large LDOS is due to SPP modes from the first layer. In figure 6 we also plot the spectral heat transfer coefficient (SHTC) \( H(\omega, z) \) which describes the spectral heat flux (also called super-Planckian radiation) between two hyperbolic multilayer structures as defined in equation (4) in [38]. It is apparent that the deviations found in the LDOS appear in the SHTC as well, so that the range of validity of the EMT is in this case the same as for the LDOS.

Now, let us see how the results change when using Ge as topmost layer. To this end, we replot figures 5 and 6 for the same structure, but with Ge on its top, in figures 7 and 8. As could be expected from the discussion for the reflection coefficient the exact results for the LDOS and the SHTC are now much better described by the EMT. In particular, we do not find an appreciable surface mode contribution within the hyperbolic bands.

Finally, in order to answer the question of how large the contribution of the hyperbolic modes to the LDOS is, we show in figure 9 the Bloch mode contribution to the LDOS normalized to the total LDOS for both cases: in (i) Ge is the topmost layer and in (ii) SiC is the topmost layer. For \( z = 500 \) nm it can be seen that for both cases the predictions of the Bloch mode contributions to the LDOS are the same. We find that the Bloch mode contributions within the band \( \Delta_1 \) are negligible as well as in the metallic region where \( \epsilon_\parallel < 0 \)

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Figure 7. As in figure 5 but with Ge as topmost layer. \( f \) is in both cases the filling fraction of SiC.
Figure 9. Plot of the relative contribution of the Bloch modes to the LDOS in the $\omega$–$f$ plane for different distances $z$ above the SiC/Ge bilayer structure, calculated with the exact S-matrix method. On the left hand side, we choose Ge as topmost layer and on the right hand side we choose SiC as topmost layer. In both cases $a = 100$ nm. $f$ is in both cases the filling fraction of SiC.

and $\epsilon_\perp < 0$ (see figure 10). On the other hand the LDOS within the band $\Delta_2$ is dominated by the hyperbolic modes. The low Bloch mode contribution in $\Delta_1$ can be explained by the fact that the hyperbolic modes for $\Delta_1$ only contribute for wavevectors larger than $|\sqrt{\epsilon_\perp} \omega/c|$. By studying figures 3(c) and 4(c) one can see that for our structures this means that $\kappa a > 0.25$. By assuming that the dominant evanescent contributions in the near-field stem from $\kappa \approx z$ we find that the hyperbolic modes within this band only contribute for $z < 4a = 400$ nm. Note that for $z = 500$ nm within the frequency band $\Delta_1$ the main evanescent contribution is due to nonresonant SPP modes, as can be observed in the reflection coefficient in figures 3(c) and 4(c) for $ka < 0.25$. Now, for $z = 200$ nm the hyperbolic Bloch modes within the band $\Delta_1$ start to contribute significantly. However, which is important, both structures do not show the same Bloch mode contributions anymore, since within the band $\Delta_2$ in figure 9(d) the resonant SPP modes of the topmost SiC-layer start to dominate the LDOS. For a distance $z = 50$ nm, i.e. for distances smaller than the period of the multilayer structures, the Bloch mode contributions to the LDOS are in both structures even more different due to the resonant SPP mode contribution within the hyperbolic bands, but also due to quenching within the effective dielectric regions. Hence, for multilayer hyperbolic structures the choice of a passive topmost layer is preferable when aiming for a large LDOS or SHTC, due to hyperbolic modes within the near-field regime.

In this work we have shown that even for distances larger than the lattice period the EMT does not provide a correct physical picture of the multilayer structure when localized modes such as surface polaritons are present. We have shown that the usual conditions for the validity of the EMT, $a \ll \lambda$ and $ka \ll 1$, have to be augmented by the condition that the distance $z$ at which the electromagnetic field is investigated must be larger than the penetration length of the SPP modes of the topmost layer into the vacuum region. Further, we have shown that the hyperbolic bands estimated by the EMT are not purely hyperbolic in nature when the topmost layer supports surface modes. This limits the validity of the EMT even for estimating the hyperbolic regions. Finally, we have shown how the unwanted surface mode contribution can be suppressed when using a passive topmost layer.

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