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Optical negative-index response of nanoscale metamaterials

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The significant limitations to optimization of left-handed metamaterials are currently inspiring attempts to devise alternative approaches to achieve a negative-index response.

Negative-index or left-handed metamaterials (LHMs, i.e., those with both electrical permittivity and negative magnetic permeability, leading to a negative index of refraction[1]) are a subject of continuously increasing research interest. This primarily originates from their novel and unique electromagnetic (EM) properties, such as opposite phase and energy velocity, negative refraction, opposite Doppler effect and Čerenkov radiation, and the possibility of achieving subwavelength-resolution imaging (superlensing) with planar samples.[2] These unique properties, in particular the superlensing possibility, have led to intense research efforts to develop such materials operating in the optical region of the EM spectrum. (Most of today's LHMs operate at microwaves.) These efforts have led to various optical metamaterial structures.[3,4] Although they constitute very interesting first developments, they are far from functional. In addition to suffering from very high losses, they are typically composed of only a single functional layer, demonstrating that the search for optimized and functional optical LHMs is far from complete.

Most current LHMs involve metallic components, but metal properties at microwaves are drastically different from those in the optical regime. Consequently, the problem of achieving optical LHMs through the common microwave approach of combining metallic negative-permittivity and permeability elements[5] is much more than a straightforward scaling problem. It involves great theoretical and experimental challenges.

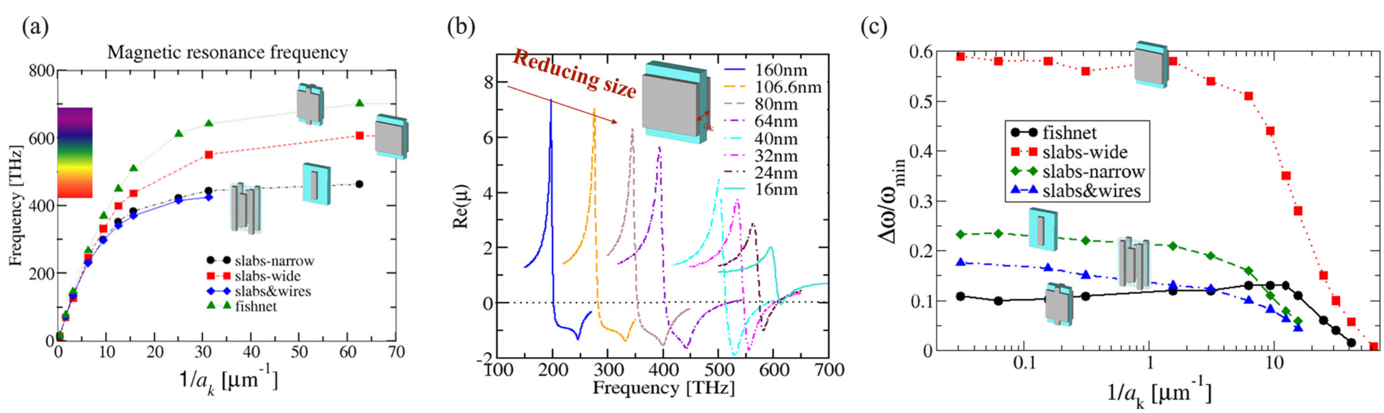

Figure 1. *Four designs for achieving optical negative-permeability response (unit cells are shown next to their corresponding curves): pairs of narrow parallel slabs, pairs of wide slabs, narrow slabs combined with continuous wires, and wide slabs connected with continuous wires (known as a fishnet). The designs are scaled uniformly from the milli- to nanometer scale. (a) Scaling of magnetic-resonance frequency versus unit-cell thickness (a_k). Although the magnetic-resonance frequency scales in inverse proportion to the unit-cell size on larger scales, at nanometer scales it saturates to a constant value, depending on the design. (b) Permeability resonance, Re(μ), as a function of frequency for the pair of wide slabs and for various unit-cell sizes. Weakening of permeability resonance in smaller-scale structures is illustrated. (c) Relative bandwidth, $\Delta\omega/\omega$, of the negative permeability band (bandwidth divided by minimum frequency for which μ is negative) as a function of unit-cell thickness. Although this relative bandwidth remains constant on larger scales (and, thus, lower frequencies), it approaches zero for nanometer-scale structures. In the simulations presented here, the metal and dielectric possess the parameters of aluminum and glass, respectively.*[7–9]

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Experimental challenges primarily arise from the inefficiency of current fabrication methods. These methods,[6] most commonly e-beam and focused ion-beam lithography, are expensive and time consuming. It is also very difficult to produce complicated patterns, large samples, and/or isotropic 3D structures, as is sometimes required, e.g., for superlensing. New fabrication approaches have recently been employed to overcome these limitations. Direct laser writing (combined with metalization) and nanoimprint lithography are the most promising among these.

An important theoretical challenge arises from the high losses in optical LHMs, caused by the high metal-induced losses in the optical regime, combined with the resonant character of the structure's response. This implies long-term interaction of the structure with the incoming EM field. Various methods of overcoming losses have been proposed. These include design optimization (to minimize the field overlap with the metallic components), a search for new metal compounds that may be optimized relative to silver (the state-of-the-art metal for fabrication of current optical LHMs), proper gain-media incorporation into metamaterials (to compensate for losses), and use of coupling approaches (e.g., electromagnetically induced transparency) to modify the resonant response.

We recently showed[7–9] that losses do not solely constitute the primary limitation toward achieving optical negative-index response. An even more important fundamental limitation arises from the inductance of the current-carrying electrons, a consequence of their finite mass. This inductance does not allow electrons to instantaneously follow high-frequency motions, as dictated by optical fields, leading to serious changes in the response of optical LHMs (relative to microwave metamaterials) and significant performance limitations.

Figure 1 provides an illustration of these issues. Magnetic-resonance-frequency saturation (i.e., saturation of the onset frequency of the negative-permeability response) in the optical regime is presented in Figure 1(a). At microwaves, this frequency is inversely proportional to the structure's length scale. Panels (b) and (c) show weakening of the magnetic-permeability (μ) resonance, until μ stops approaching negative values, and vanishing of the negative-permeability bandwidth, respectively. At microwaves, the relative bandwidth (bandwidth divided by resonance frequency) of the negative-permeability band remains constant by uniformly scaling the structures.

We verified, based on detailed numerical simulations,[7–9] that both of these effects are completely independent of ohmic losses. This property, along with the primary characteristics of the optical negative permeability and index response, can be predicted by simple analytical models. These include describing the magnetic resonator (i.e., the LHM component responsible for the negative-μ response) as an effective resistor-inductor-capacitor (RLC) circuit, using simplified formulas for the capacitance, inductance, and resistance of the circuit, and using the *frequency-dependent* free-electron conductivity model for metal conductivity instead of the real, frequency-independent conductivity appropriate for microwaves. Such an all-inclusive model can predict the primary geometric and material factors that determine the magnetic response of optical negative permeability and LHMs, and lead to simple design rules for optimization of such metamaterials.[7–9]

Applying and analyzing such a model can lead to the conclusion that one of the most important factors determining the response of high-frequency LHMs is the *plasma frequency* of the metal involved in metamaterial development, which must be as high as possible to ensure large negative-permeability bandwidth and a high saturation value for the magnetic-resonance frequency. It also illustrates the important role of geometry, since this determines the structure's effective inductance and capacitance. Designs of low capacitance favor optimized high-frequency response. Low inductance, although also favoring higher-frequency response, leads to smaller operational bandwidths and larger losses.

The effective RLC model illustrates that another factor that determines the strength of the permeability resonance (which is responsible for the negative-permeability response) in an LHM is the metal's loss factor. The latter should be as low as possible to effect a strong response with negative μ values.

Both the finite plasma frequency and high loss factors of current metallic components lead to unavoidable deterioration of the negative-permeability response in current optical metamaterials. These almost unavoidable limitations are currently inspiring attempts, including by our team, to devise alternative approaches to achieve an optical negative-index response (beyond the combined negative-permittivity and permeability approach), e.g., involving chirality.[10,11] Further developments include alternative materials with low effective electron mass (and thus high plasma frequency), such as graphene, to achieve optimized high-frequency, negative-index structures. This can lead to novel optimized optical LHMs, and can also open up new research directions leading to new possibilities and surprises, an increasingly common occurrence in the metamaterials field.

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