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LETTER TO THE EDITOR

Multilayer structures as negative refractive and left-handed materials

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Abstract
We examine multilayer structures as negative refractive index and left-handed materials, and find that for one polarization there is a wide range (≈90°) of incident angle within which negative refraction will occur. This comes about because the group velocity and the Poynting vector have a large component parallel to the layers, no matter what the angle of incidence of the incoming radiation is. This behaviour in turn comes from the large anisotropy of the phase velocities. If one of the components is a ferromagnetic metal, the system can be a left-handed material above the ferromagnetic resonance frequency.

There has been much recent interest in left-handed and negative refractive index materials. Left-handed materials are materials in which the direction of energy flow of an electromagnetic wave is opposite to the wavevector. In negative refractive material the component of the average Poynting vector of an electromagnetic wave tangential to the interface changes sign after refraction. The original idea focuses on materials with negative dielectric constants ϵ and negative magnetic permeabilities μ [1, 2]. *Isotropic* left-handed materials with negative dielectric constants and magnetic susceptibilities are also negative refractive index materials. The first experimental demonstration of left-handed materials is on a system of split ring resonators and wires [3]. *Anisotropic* materials that exhibit negative refraction are not necessarily left handed [4–6]. The requirements for left-handed materials are also different. Recently, Zhang and co-workers [7] demonstrated negative refraction at the interface of a crystal with uniaxially anisotropic positive definite susceptibilities. This negative refraction occurs when the angle of the incoming radiation is within a range θ_u from the surface normal. The magnitude of the range θ_u depends on the degree of anisotropy [8].

In this paper we examine multilayer structures as negative refractive index and left-handed materials. Our motivation is twofold. (1) In multilayer structures, the propagation of electromagnetic waves can be exactly analytically calculated. This will provide us with some assessment of different approximations that has been used in this area. (2) The system of metal–insulator multilayers is highly anisotropic: it is a metal for current flows along the planes and an insulator for current flows perpendicular to the planes. Thus this system may exhibit a wide range θ_u of incident angle within which negative refraction will occur. We find that for a polarization with the macroscopic magnetic field H perpendicular to the wavevector and parallel to the layers, there is a large range of incident angles that is close to 90° within which the radiation is refracted negatively. This comes about because the group velocity and the Poynting vector have a large component parallel to the layers, no matter what the angle of incidence of the incoming radiation is. This anisotropic group velocity in turn comes from the large anisotropy of the phase velocities. When the wavevector is parallel (perpendicular) to the layers, the square of the phase velocity is inversely proportional to $\langle\mu\rangle_{\mathrm{a}}\langle\epsilon\rangle_{\mathrm{h}}$ ($\langle\epsilon\rangle_{\mathrm{a}}\langle\mu\rangle_{\mathrm{a}}$), where the angular brackets with subscripts a, h stand for the arithmetic and the harmonic mean respectively. For example: $\langle\epsilon\rangle_{\mathrm{a}} = c_m\epsilon_m + c_i\epsilon_i$, $1/\langle\epsilon\rangle_{\mathrm{h}} = c_m/\epsilon_m + c_i/\epsilon_i$ where c_j stands for the volume fraction of component j. $\langle\epsilon\rangle_{\mathrm{a}}$ is of the same order of magnitude of the metal dielectric constant whereas $\langle\epsilon\rangle_{\mathrm{h}}$ is of the order of magnitude of the insulator dielectric constant. These two are very different. The different averages of the dielectric constant obtained here in the long wavelength limit is very similar to the ‘form birefringence’ discussed by Born and Wolf [9]. The form birefringence focuses on the dielectric constants only. Here we have included the magnetic susceptibility at the same time. Our system exhibits negative refraction over a *wide* frequency range from the visible to microwave. Current systems exhibit negative refraction over a limited range of frequencies near the microwave and suffer heavy losses (>20 dB). Our system offers an alternative way to tackle this problem.

We begin by considering the propagation of electromagnetic waves in the multilayer structure consisting of periodic arrays of two materials of thicknesses d_m, d_i with $d = d_m + d_i$. The dielectric constant and the magnetic permeability of the two components are denoted by ϵ_m, μ_m (ϵ_i, μ_i). There are two types of eigenstates for Maxwell’s equation: the H (E) polarization where the macroscopic magnetic field H (electric field E) is perpendicular to the wavevector and parallel to the layers. Our goal is to derive the dispersion relationships of the radiation for these two polarizations in the long wavelength limit. For the H (E) polarization we denote the direction of $\mathbf{H}$ ($\mathbf{E}$) as the y direction and the normal to the multilayers as the x direction. Any wavevector $\mathbf{k}$ can be decomposed into a component perpendicular to the planes and a component parallel to the planes. The wavevector is in the xz plane with $\mathbf{k} = (k_x, 0, k_z)$. There is no y component because the wavevector is perpendicular to the direction of $\mathbf{H}$ ($\mathbf{E}$). The frequency of the radiation will be denoted by ω. We define a ‘vacuum wavevector’ $k_0 = \omega/c$ where c is the speed of light.

To calculate the dispersion we solve Maxwell’s equation in each component separately. The solution is then matched across the boundary [10, 11]. The solution of Maxwell’s equation in each region $j = m, i$ can be written in separable form as $E_{zj} = V_j(x)\exp(\mathrm{i}k_z z)$, $H_{yj} = X_j(x)\exp(\mathrm{i}k_z z)$ for the H polarization and $H_{zj} = V_j(x)\exp(\mathrm{i}k_z z)$, $E_{yj} = X_j(x)\exp(\mathrm{i}k_z z)$ for the E polarization. The wavevector k_z, the component of the wavevector parallel to the planes, is the same in both regions. The functions V and X are linear combinations of plane wave solutions. $X_j = A_j\cos(p_j x') + B_j\sin(p_j x')$, $V_j = \mathrm{i}[-p_j A_j\sin(p_j x') + p_j B_j\cos(p_j x')]/(k_0\tau_j)$ where $p_j = (\epsilon_j\mu_j k_0^2 - k_z^2)^{0.5}$. $\tau_j = \epsilon_j$ ($-\mu_j$) for the H (E) polarization. $x' = x$ for $0 < x < d_m$; $x' = x - d_m$ for $d_m < x < d_i + d_m$. The constant coefficients A_j and B_j can be determined from the continuity of the tangential components of E and H across the boundaries and the ‘periodic boundary condition’: $E(x + d) = \exp(\mathrm{i}k_x d)E(x)$, $H(x + d) = \exp(\mathrm{i}k_x d)H(x)$. Across the first interface, we get from the continuity conditions

$$\begin{bmatrix} X_m(x = d_m) \\ V_m(x = d_m) \end{bmatrix} = \mathbf{T}(p_m, d_m, \tau_m)\begin{bmatrix} X_m(x = 0) \\ V_m(x = 0) \end{bmatrix}$$

where

$$\mathbf{T}(p,d,\tau)=\begin{bmatrix}\cos(pd) & -\mathrm{i}\sin(pd)\tau k_0/p\\ -\mathrm{i}\sin(pd)p/\tau k_0 & \cos(pd)\end{bmatrix}.$$

From the ‘periodic boundary condition’, we get

$$\begin{bmatrix}X_i(d)\\ V_i(d)\end{bmatrix}=\exp[\mathrm{i}dk_x]\begin{bmatrix}X_i(0)\\ V_i(0)\end{bmatrix}.$$

Multiplying the above two equations, we obtain an eigenvalue problem.

$$\begin{bmatrix}X_i(0)\\ V_i(0)\end{bmatrix}=\exp[-\mathrm{i}dk_x]\mathbf{T}(p_m,d_m,\tau_m)\mathbf{T}(p_i,d_i,\tau_i)\begin{bmatrix}X_i(0)\\ V_i(0)\end{bmatrix}.$$

Simplifying the algebra[4], we obtain finally the eigenvalue equation

$$\cos(k_xd)=\cos\phi_i\cos\phi_m-0.5[\kappa p_i/p_m+p_m/(p_i\kappa)]\sin\phi_i\sin\phi_m \tag{1}$$

where $\phi_j = p_jd_j$, $\kappa = \tau_m/\tau_i$. The corresponding eigenvector is given by $A_m = 1$, $B_m = \mathrm{i}W_0\tau_mk_0/p_m$ where $W_0 = [\exp(\mathrm{i}k_xd) - M]/N$. $M = \cos(p_md_m)\cos(p_id_i) - p_m\tau_i\sin(p_id_i)\sin(p_md_m)/(\tau_mp_i)$, $N = \mathrm{i}k_0[\cos(p_md_m)\sin(p_id_i)\tau_i/p_i + \cos(p_id_i)\sin(p_md_m)\tau_m/p_m]$. $A_i = U_1$, $B_i = \mathrm{i}W_1\tau_ik_0/p_i$ where $U_1 = \cos(p_md_m) + \mathrm{i}W_0\tau_mk_0\sin(p_md_m)/p_m$, $W_1 = V_0\cos(p_md_m) + \mathrm{i}p_m\sin(p_md_m)/(\tau_mk_0)$.

We next examine these results in the long wavelength limit with $p_jd_j \ll 1$. Using the approximation $\cos(x)\approx 1-x^2/2$, $\sin(x)\approx x$ we get from equation (1) after some algebra

$$\begin{aligned}&k_z^2(d_i\sqrt{\kappa}+d_m/\sqrt{\kappa})+(k_xd)^2/(d_i/\sqrt{\kappa}+d_m\sqrt{\kappa})\\ &\qquad=k_0^2(\mu_i\epsilon_id_i\sqrt{\kappa}+\mu_m\epsilon_md_m/\sqrt{\kappa}).\end{aligned}\tag{2}$$

Putting in the expression for κ and simplifying, we get for the H polarization

$$k_z^2/\langle\epsilon\rangle_{\mathrm{h}}+k_x^2/\langle\epsilon\rangle_{\mathrm{a}}=k_0^2\langle\mu\rangle_{\mathrm{a}} \tag{3}$$

where the angular brackets with a subscript a stand for the arithmetic mean: $\langle\epsilon\rangle_{\mathrm{a}} = (d_m\epsilon_m + d_i\epsilon_i)/d$, $\langle\mu\rangle_{\mathrm{a}} = (d_m\mu_m + d_i\mu_i)/d$. Similarly, angular brackets with a subscript h stand for the harmonic mean: $1/\langle\epsilon\rangle_{\mathrm{h}} = (d_m/\epsilon_m + d_i/\epsilon_i)/d$.

For the E polarization, one interchanges ϵ with μ. From equation (6), we get the long wavelength dispersion:

$$k_z^2/\langle\mu\rangle_{\mathrm{h}}+k_x^2/\langle\mu\rangle_{\mathrm{a}}=k_0^2\langle\epsilon\rangle_{\mathrm{a}}. \tag{4}$$

The system is also anisotropic. The real part of $\langle\epsilon\rangle_{\mathrm{a}}$ is negative. In this long wavelength limit the corresponding eigenvector becomes

$$X_m=\cos(p_mx)+\mathrm{i}k_x\tau_m\sin(p_mx)/[p_m\langle\tau\rangle_{\mathrm{a}}], \tag{5}$$

$$X_i=\cos(p_ix')+\mathrm{i}k_x\tau_i\sin(p_ix')/[p_i\langle\tau\rangle_{\mathrm{a}}]. \tag{6}$$

We next look at the Poynting vector of the system. We first discuss the case of the H polarization. We get

$$\vec{H}_j=H_0\vec{\mathbf{y}}\mathrm{e}^{\mathrm{i}k_zz-\mathrm{i}\omega t}X_j(x), \tag{7}$$

$$\vec{E}_j=H_0\left[\frac{k_z}{k_0\epsilon_j}\vec{\mathbf{x}}X_j(x)+\mathrm{i}\frac{1}{k_0\epsilon_j}\vec{\mathbf{z}}X_j'(x)\right]\mathrm{e}^{\mathrm{i}k_zz-\mathrm{i}\omega t}. \tag{8}$$

[4] Define a matrix $U = T(p_m, d_m, \tau_m)T(p_i, d_i, \tau_i)$. One can show by direct computation that the determinant of the matrix U is unity. The eigenvalue equation $\det(U-\lambda)=0$ reduces to a quadratic equation $\lambda^2-2z\lambda+1=0$ where z is the right-hand side of equation (1). The solution of this equation is $\lambda=z\pm(z^2-1)^{0.5}$. If one calls $z=\cos kl$, then $\lambda=\exp(\pm\mathrm{i}kl)$, as claimed.

The corresponding Poynting vector $\mathbf{S}_j = \mathbf{E}_j \times \mathbf{H^*}_j$ is given by (as usual [12], it is the real part of this expression that is of interest)

$$\vec{S}_j = H_0^2\left(-\frac{\mathrm{i}X_j^*X_j'}{2k_0\epsilon_j}\vec{\mathbf{x}} + \frac{|X_j|^2k_z}{2k_0\epsilon_j}\vec{\mathbf{z}}\right),$$

X_j is a function of the spatial variable x. We calculate the mean Poynting vector by averaging expressions involving X_m, X_m' (X_i, X_i') in the interval $0 < x < d_m$ ($d_m < x < d + m + d_i$). From equations (5) and (6), we can calculate the averages of expressions involving the function X: $\langle |X|^2\rangle = 1$, $\langle X^*X'\rangle = \mathrm{i}k_x\tau/\langle\tau\rangle_{\mathrm{a}}$.

The Poynting vector in the corresponding region is

$$\vec{S}_j = H_0^2\left(\frac{k_x}{2k_0\langle\epsilon\rangle_{\mathrm{a}}}\vec{\mathbf{x}} + \frac{k_z}{2k_0\epsilon_j}\vec{\mathbf{z}}\right).$$

Averaging over the two types of layers, we get

$$\vec{S} = H_0^2\left(\frac{k_x}{2k_0\langle\epsilon\rangle_{\mathrm{a}}}\vec{\mathbf{x}} + \frac{k_z}{2k_0\langle\epsilon\rangle_{\mathrm{h}}}\vec{\mathbf{z}}\right). \tag{9}$$

This Poynting vector is parallel (antiparallel) to the normal of the constant ω contour, $\partial\omega/\partial\mathbf{k}$, if $\langle\mu\rangle_{\mathrm{a}}$ is positive (negative). If the imaginary parts of the susceptibilities are small so that the wavevector is mostly real, the dot product of the wavevector and the Poynting vector is given by

$$\vec{k}^* \cdot \vec{S} \approx \tfrac{1}{2}k_0\langle\mu\rangle_{\mathrm{a}}|H_0|^2. \tag{10}$$

Thus if we can find a material with a negative average $\langle\mu\rangle_{\mathrm{a}}$, Real$[\mathbf{k}^* \cdot \mathbf{S}] < 0$, the system will be left handed. This may be achievable with ferromagnetic materials above the ferromagnetic resonance frequency; we shall come back to this point later.

We next discuss the case of the E polarization. The electric and magnetic fields are given by

$$\vec{E}_j = E_0\vec{\mathbf{y}}\mathrm{e}^{\mathrm{i}k_zz-\mathrm{i}\omega t}X_j(x), \tag{11}$$

$$\vec{H}_j = E_0\left[-\frac{k_z}{k_0\mu_j}\vec{\mathbf{x}}X_j(x) - \mathrm{i}\frac{1}{k_0\mu_j}\vec{\mathbf{z}}X_j'(x)\right]\mathrm{e}^{\mathrm{i}k_zz-\mathrm{i}\omega t}. \tag{12}$$

The Poynting vector is now given by

$$\vec{S}_j = E_0^2\left(-\frac{\mathrm{i}X_j^*X_j'}{2k_0\mu_j}\vec{\mathbf{x}} + \frac{|X_j|^2k_z}{2k_0\mu_j}\vec{\mathbf{z}}\right).$$

Substituting in the average of the function X_j, we get

$$\vec{S}_j = E_0^2\left(\frac{k_x}{2k_0\langle\mu\rangle_{\mathrm{a}}}\vec{\mathbf{x}} + \frac{k_z}{2k_0\mu_j}\vec{\mathbf{z}}\right).$$

Averaging over the two components we get

$$\vec{S} = |E_0|^2\left(\frac{k_x}{2k_0\langle\mu\rangle_{\mathrm{a}}}\vec{\mathbf{x}} + \frac{k_z}{2k_0\langle\mu\rangle_{\mathrm{h}}}\vec{\mathbf{z}}\right), \tag{13}$$

$$\vec{k} \cdot \vec{S} = \tfrac{1}{2}k_0\langle\epsilon\rangle_{\mathrm{a}}|E_0|^2. \tag{14}$$

The Poynting vector is again anisotropic. The same results are obtained if one approximates the multilayer system as an anisotropic homogeneous system [5]. If the imaginary part $\langle\epsilon\rangle_{\mathrm{a}}$ is small $\mathbf{k}\cdot\mathbf{S} < 0$. We next consider the multilayer structure of metals and insulators as a negative refractive index material.

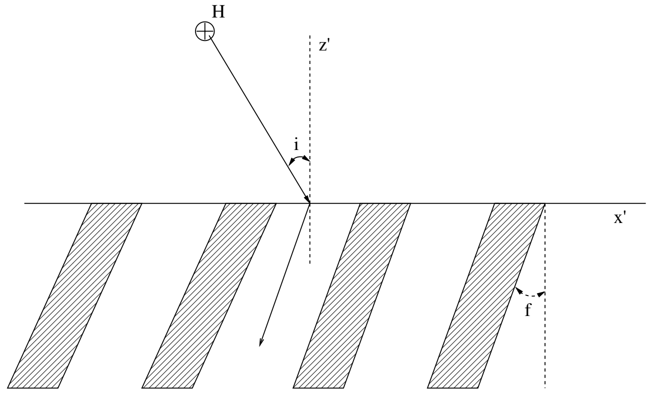

Figure 1. The geometry of the refraction from the multilayer.

The geometry we have in mind is shown in figure 1. The plane of incidence is in the $x'z'$ plane. The multilayers are at an angle f with respect to the $y'z'$ plane. The incident radiation comes in at an angle i with respect to the interface normal which is assumed to be in the z' direction. This geometry is a little bit different from the conventional arrangement with the interface parallel to the layers. We are interested in the regime where the spacing is much less than a wavelength and the wave is not confined in a single insulating region confined by the metallic layers. Thus our structure is not operated as a waveguide. First consider the H polarization. At frequencies of the order of GHz, the dielectric constant of the metal is much larger than that of the insulator. $\langle\epsilon\rangle_{\mathrm{h}} \approx \epsilon_i d/d_i$ whereas $\langle\epsilon\rangle_{\mathrm{a}} \approx \epsilon_m d_m/d$. *This result is valid even when the imaginary parts of the susceptibilities of the metal are large*. After refraction, the x' component of the wavevector, $q_{x'}$ is equal to the x' component of the incoming wavevector $k_{x'} = k_0 \sin i$. The component of the wavevector of the refracted radiation parallel to the layers is given by $q_z = q_{z'} \cos f - q_{x'} \sin f$. The component normal to the layers is given by $q_x = q_{z'} \sin f + q_{x'} \cos f$. The frequencies of the incoming and refracted radiation are the same. Since $|\langle\epsilon\rangle_{\mathrm{a}}| \gg \langle\epsilon\rangle_{\mathrm{h}}$, we get from equation (7) $q_z = k_0(\langle\mu\rangle_{\mathrm{a}}\langle\epsilon\rangle_{\mathrm{h}})^{0.5}$. Again, because $|\langle\epsilon\rangle_{\mathrm{a}}| \gg \langle\epsilon\rangle_{\mathrm{h}}$ from equation (13) the Poynting vector is given by

$$\vec{S} \approx H_0^2 \frac{q_z}{2k_0\langle\epsilon\rangle_{\mathrm{h}}}\vec{\mathbf{z}}. \tag{15}$$

Thus no matter what angle the incident radiation comes in, it will always be refracted along the direction of the multilayers. There is very strong negative refraction.

For the E polarization, since $\langle\mu\rangle_{\mathrm{a}} \gg \langle\mu\rangle_{\mathrm{h}}$, we get from equation (17)

$$\vec{S} \approx |E_0|^2 \frac{k_z}{2k_0\langle\mu\rangle_{\mathrm{h}}}\vec{\mathbf{z}}.$$

Again, the wave will be negatively refracted along the layer direction. In general $\langle\epsilon\rangle_{\mathrm{a}}$ will have a significant imaginary part. From equation (8), we expect $k_z \approx k_0(\langle\epsilon\rangle_{\mathrm{a}}\langle\mu\rangle_{\mathrm{h}})^{0.5}$ to possess a significant imaginary part. This mode will be heavily damped.

We close with a discussion of how to make $\langle\mu\rangle_{\mathrm{a}}$ negative. The obvious choice is to use a ferromagnetic metal as one of the components of the multilayer system [14]. When the magnetization is aligned along the z direction, the magnetic susceptibility of a ferromagnet is a tensor given by [13]

$$\hat{\mu}_{\mathrm{F}} = \begin{bmatrix} \mu_d & -\mathrm{i}\mu' & 0 \\ \mathrm{i}\mu' & \mu_d & 0 \\ 0 & 0 & 1 \end{bmatrix} \tag{16}$$

where

$$\mu_d = 1 + \frac{\omega_m(\omega_0 - \mathrm{i}\alpha\omega)}{(\omega_0 - \mathrm{i}\alpha\omega)^2 - \omega^2}, \tag{17}$$

$$\mu' = -\frac{\omega_m\omega}{(\omega_0 - \mathrm{i}\alpha\omega)^2 - \omega^2}. \tag{18}$$

Here $\omega_0 = \gamma|\vec{H}_0|$ is the ferromagnetic resonance frequency, H_0 is the effective magnetic field in magnetic particles and may be a sum of the external magnetic field, the effective anisotropy field and the demagnetization field; $\omega_m = \gamma|\vec{M}_0|$, with γ the gyromagnetic ratio and M_0 the saturation magnetization of magnetic particles; α is the magnetic damping coefficient; and, finally, ω is the frequency of incident electromagnetic waves. It is still possible to solve Maxwell's equations for the multilayer system analytically. The results are algebraically complicated and not very illuminating physically. Here we consider the case when the remanent magnetization of the ferromagnet is zero. The system consists of domains with the magnetization forced by the shape anisotropy to lie in the yz plane but otherwise randomly oriented. For frequencies of the order of GHz, the domain size is usually much less than the wavelength. The magnetic susceptibility can be obtained by averaging $\hat{\mu}_\mathrm{F}$ over the different orientations of the magnetizations of the domains. The resulting magnetic susceptibility becomes diagonal but anisotropic:

$$\hat{\mu}_{\mathbf{M}=0} = \begin{bmatrix} \mu_d & 0 & 0 \\ 0 & \mu_{yz} & 0 \\ 0 & 0 & \mu_{yz} \end{bmatrix} \tag{19}$$

where $\mu_{yz} = (\mu_d + 1)/2$. It is still possible to solve Maxwell's equation analytically. For the H polarization, the magnetic susceptibility μ_m is now replaced by μ_{yz}. Above the ferromagnetic resonance frequency ω_0, if μ_d becomes negative enough that $\langle\mu\rangle_\mathrm{a}$ is also negative, then the system can be considered a left-handed material.

For the E polarization,

$$\vec{E} = E_0\vec{\mathbf{y}}\mathrm{e}^{\mathrm{i}k_z z - \mathrm{i}\omega t}X(x), \tag{20}$$

$$\vec{H} = E_0\left[-\frac{k_z}{k_0\mu_d}\vec{\mathbf{x}}X(x) - \mathrm{i}\frac{1}{k_0\mu_{yz}}\vec{\mathbf{z}}X'(x)\right]\mathrm{e}^{\mathrm{i}k_z z - \mathrm{i}\omega t}. \tag{21}$$

Now $p_m = (k_0^2\epsilon_m\mu_{yz} - k_z^2\mu_{yz}/\mu_d)^{0.5}$, $\tau_m = \mu_{yz}$. The functional form for the dispersion, equation (8), remains the same except now the different averages of the magnetic susceptibilities involve different components of the tensor: $1/\langle\mu\rangle_\mathrm{h} = c_i/\mu_i + c_m/\mu_d$; $\langle\mu\rangle_\mathrm{a} = c_i\mu_i + c_m\mu_{yz}$. The conclusions reached previously remain qualitatively unchanged.

In conclusion, we propose that a multilayer structure can refract negatively. The physical reason is that the group velocity is very anisotropic. In this paper we have assumed that $k_jd_j \ll 1$. Typically, in multilayers d_j can easily be made to be of the order of 10 Å. k_j is of the order of $2\pi(\epsilon)^{0.5}/(\text{wavelength})$. For microwaves with wavelengths of the order of millimetres (10^6 Å), ϵ is of the order of 10^5 [15]. No matter what the angle of incidence is, the largest value of $kd \approx 10^{-2}$. For infrared radiation, the wavelength is of the order of a micron (10^4 Å) and ϵ is of the order of 10. Again, for all possible angles of incidence, the largest possible $kd \approx 10^{-3}$. Thus at different frequencies our condition can be easily satisfied. The transmission in these types of systems can be estimated from the reflectivity previously calculated [11]. In the infrared, for a Cu–Ge system, the reflectivity can be made as low as 10%. For a thin enough system, a high transmission of 90% can be obtained. Typical interface roughness is of the order of ångströms whereas the wavelengths of interest are more than thousands of ångströms. The interface roughness is much less than a wavelength. There are other structures that can work under the same philosophy. An example is an array of parallel cylinders (wires). For current flow parallel (perpendicular) to the wires, the system behaves like a metal (an insulator). We have performed preliminary calculations, that suggest a very similar scenario for that case as well.

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