Coupled Plasmons and Resonant Effective Permeability of Metal-Dielectric-Metal Nanosandwich Assemblies

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Periodic assemblies of in-tandem pairs of metallic nanodisks separated by a dielectric spacer, so-called metal-dielectric-metal nanosandwiches, constitute a novel class of photonic metamaterials with intriguing optical properties. When two metallic nanodisks are brought into strong coupling in a sandwich-like configuration, plasmon hybridization results into a symmetric resonant mode, with the dipole moments of the nanodisks oscillating in phase, and an antisymmetric resonant mode, with the dipole moments oscillating with opposite phase. While the symmetric resonance has an electric dipolar character, the antisymmetric one is associated with a loop-like current in the nanodisk pair and thus a dipole magnetic moment [1]. Two- and three-dimensional structures of such photonic metamolecules may exhibit a negative effective permeability in the region of the antisymmetric resonance, at visible and near-infrared frequencies [2], which is an essential ingredient in the design of negative-index metamaterials. Compared to pairs of rods or of cut-wires, the optical behavior of disk pairs is more isotropic because the latter are invariant under rotation about their axis. It is worthnoting that, contrary to the formation of bonding and antibonding electron orbitals in diatomic molecules, in a metal-dielectric-metal nanosandwich, the low-frequency hybrid plasmonic mode is antisymmetric and the high-frequency one is symmetric. This apparently counter-intuitive situation can be understood as follows. Charge oscillations associated with an electric-dipole plasmon mode in a single metallic nanodisk are sustained by restoring forces acting on the collectively displaced conduction-band electrons. In an in-tandem pair of such nanodisks, charge distribution leads to reduction of the appearing restoring forces in the configuration of the antisymmetric mode and enhancement in the case of the symmetric mode. Consequently, the eigenfrequency of the antisymmetric mode is lowered and that of the symmetric mode is raised, as shown schematically in Fig. 1. The situation is reversed if the two nanodisks are on the same plane.

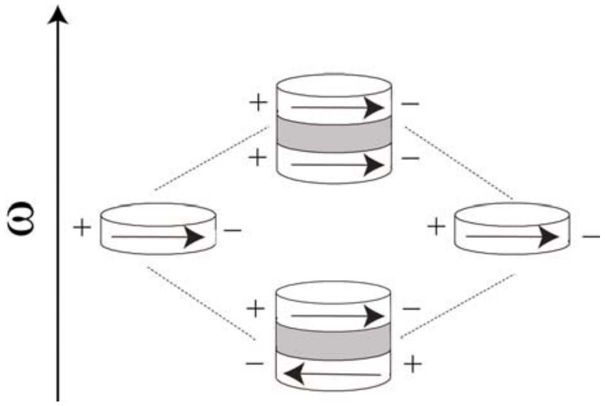

Fig.1. A schematic description of plasmon hybridization in a metal-dielectric-metal nanosandwich.

In the present communication, we report on the effective magnetic permeability of two- and three-dimensional periodic structures of metal-dielectric-metal nanosandwiches by means of full electrodynamic calculations using the extended layer-multiple-scattering method [3]. This method provides a versatile and efficient computational framework for fast and accurate calculations of the optical properties of complex inhomogeneous systems consisting of successive, possibly different, layers of scatterers arranged with the same two-dimensional periodicity. The properties of the individual scatterers enter only through the corresponding T matrix which, for scatterers of arbitrary shape, is calculated numerically by the extended boundary condition method. At a first step, in-plane multiple scattering is evaluated in a spherical-wave basis with the help of proper propagator functions. Subsequently, interlayer scattering is calculated in a plane-wave basis through appropriate transmission and reflection matrices. The scattering S matrix of a multilayer slab, which transforms the incident into the outgoing wave field, is obtained by combining the transmission and reflection matrices of the component layers. For a three-dimensional crystal consisting of an infinite periodic sequence of layers, Bloch theorem leads to an eigenvalue equation that gives the (complex) normal component of the Bloch wave vector, k_z, for given frequency, ω, and in-plane reduced wave vector component, $\mathbf{k}_\|$, which are (real) conserved quantities in the scattering process. The effective electromagnetic parameters of the structure, i.e., the permittivity and permeability functions of an equivalent homogeneous medium, are determined from the scattered field in the far zone by a finite slab of the structure, under plane wave illumination. At normal incidence, inverting the standard Fresnel equations, we obtain closed-form solutions for the effective refractive index, n_{eff}, and impedance, z_{eff}, in terms of the complex transmission and reflection coefficients (S -matrix retrieval) [2]. As the thickness of the slab increases, n_{eff} should converge to ck_z/ω (c is the velocity of light in vacuum), which is unambiguously deduced from the complex photonic band structure of the corresponding infinite crystal. The effective permittivity and permeability of the slab are given by $\varepsilon_{\mathrm{eff}} = n_{\mathrm{eff}}/z_{\mathrm{eff}}$ and $\mu_{\mathrm{eff}} = n_{\mathrm{eff}} z_{\mathrm{eff}}$. Obviously, $\varepsilon_{\mathrm{eff}}$ and μ_{eff} do not describe the wave field inside the actual structure where, at a given frequency, it has the form of a Bloch wave rather than a simple plane wave. However, the effective parameters must be such that these two waves obey the same dispersion relation and, therefore have the same group (and phase) velocity. This remark is of course meaningful only if there is a single dominant relevant Bloch mode at the given frequency. Moreover, in order for an effective-medium description to be applicable, the wavelength in the embedding medium must be much larger than the in-plane period of the structure. This condition ensures that there is only a single propagating mode of the scattered electromagnetic field corresponding to outgoing waves (refracted and reflected beams). All other components of the wave field (diffracted beams) are evanescent.

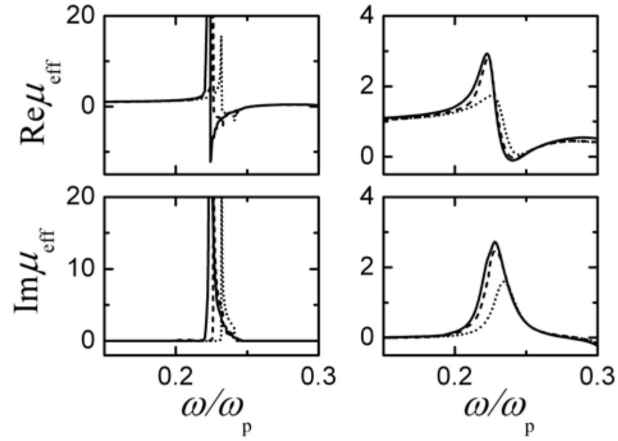

Fig. 2. Effective permeability of one- (dotted lines), two- (dashed lines) and eight- (solid lines) layers thick slabs of the structure under consideration, at normal incidence. Left: Without losses ($\gamma = 0$). Right: With losses ($\gamma = 0.025\omega_{\mathrm{p}}$).

We consider layered structures of metal-dielectric-metal nanosandwiches. In each layer, the nanosandwiches are arranged on a hexagonal lattice determined by the primitive vectors $\mathbf{a}_1 = a_0(1,0,0)$ and $\mathbf{a}_2 = a_0(1/2, \sqrt{3}/2, 0)$. We assume that the permittivity of the metallic material is described by the Drude dielectric function, $\varepsilon_{\mathrm{m}} = 1 - \omega_{\mathrm{p}}^2/\omega(\omega + i\gamma)$, where ω_{p} is the bulk plasma frequency and γ a damping factor that accounts for dissipative losses. The nanosandwiches consist of two metallic nanodisks, of radius $S = 2.5c/\omega_{\mathrm{p}}$ and thickness $h_1 = h_3 = c/\omega_{\mathrm{p}}$, separated by a silica spacer ($\varepsilon_{\mathrm{silica}} = 2.13$) of thickness $h_2 = 2c/\omega_{\mathrm{p}}$. Therefore, $h = h_1 + h_2 + h_3 = 4c/\omega_{\mathrm{p}}$ is the total thickness of the nanosandwich. The stacking sequence is defined by $\mathbf{a}_3 = (a_0/2, a_0\sqrt{3}/6, h)$. We take $a_0 = 10c/\omega_{\mathrm{p}}$. In Fig. 2 we display the retrieved μ_{eff} , for slabs one-, two- and eight-layers thick, which clearly converges with increasing slab thickness and exhibits a resonant behavior about the frequency of the antisymmetric plasmon modes. It can be seen that, even assuming that the building units of the structure are non-absorptive with purely real permittivities and permeabilities, the S -matrix retrieval method leads to non-zero imaginary part for μ_{eff} (and also for $\varepsilon_{\mathrm{eff}}$ that we don't show here) in the frequency region of the resonance. However, the retrieval procedure itself ensures that the values of $\varepsilon_{\mathrm{eff}}$ and μ_{eff} are such that the absorption of each effective slab vanishes at any frequency. In some sense, it is not possible that the effective slab complies with the strong restriction to reproduce exactly the transmission and reflection coefficients of the actual metamaterial slab, with real functions $\varepsilon_{\mathrm{eff}}(\omega)$ and $\mu_{\mathrm{eff}}(\omega)$ in the resonance region. To make this possible, one has to assume complex functions with negative $\mathrm{Im}\varepsilon_{\mathrm{eff}}$ and positive $\mathrm{Im}\mu_{\mathrm{eff}}$, i.e., some fictitious dielectric gain, which counterbalances the fictitious magnetic losses. Obviously, this occurs for given slab thickness, specific characteristics of the incident field, etc. and, therefore, $\varepsilon_{\mathrm{eff}}$ and μ_{eff} have not the meaning of inherent material parameters [4]. If absorptive losses in the metallic material are taken into account, the resonance structures become smoother and more extended in frequency while the region of negative permeability shrinks and almost disappears, as shown in Fig. 2.

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