Effective electric and magnetic properties of metasurfaces in transition from crystalline to amorphous state

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In this paper we theoretically study electromagnetic reflection, transmission, and scattering properties of periodic and random arrays of particles which exhibit both electric-mode and magnetic-mode resonances. We compare the properties of regular and random grids and explain recently observed dramatic differences in resonance broadening in the electric and magnetic modes of random arrays. We show that randomness in the particle positioning influences equally on the scattering loss from both electric and magnetic dipoles, however, the observed resonance broadening can be very different depending on the absorption level in different modes as well as on the average electrical distance between the particles. The theory is illustrated by an example of a planar metasurface composed of cut-wire pairs. We show that in this particular case at the magnetic resonance the array response is almost not affected by positioning randomness due to lower frequency and higher absorption losses in that mode. The developed model allows predictions of behavior of random grids based on the knowledge of polarizabilities of single inclusions.

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I. INTRODUCTION

Metamaterials are artificial composite materials which possess unusual electromagnetic properties not normally found in natural materials.[1] Electromagnetic properties of nanostructured metamaterials in the optical range are one of the foci of interest in modern electromagnetics.[2–7] Traditionally, metamaterials and metasurfaces composed of small individual resonant inclusions are realized as periodical arrays.[8–13] However, most recently, random or amorphous metamaterials start to attract attention, see Refs. 14–21. This is due to novel technological possibilities to manufacture amorphous structures cheaply and on a large scale, using advanced self-assembly techniques. In addition, effects of strong spatial dispersion (often undesirable) can be in some cases suppressed in disordered structures. It is generally accepted that the electromagnetic properties of both regular and random arrays of scatterers are quite similar if the distances between inclusions are electrically small. The main difference in electromagnetic response comes from scattering on the lattice inhomogeneities. This apparently results in additional loss in amorphous metamaterials, and for this reason regular metamaterial lattices have been the preferred choice if low-loss response is desired.

However, it appears that in metamaterial structures exhibiting resonant responses in several modes, the effects due to position randomness of inclusions are more complicated. In a recent paper[14] by Helgert *et al.* reflection and transmission properties of regular and random (amorphous) planar arrays of cut-wire particles were studied both numerically and experimentally. Specially introduced position disorder of individual scatterers allowed us to study the effect of distortion of periodicity on the electromagnetic response of the array. It was found that position randomness drastically affects the electromagnetic behavior at the electric resonance, but makes little impact at the array properties near the magnetic resonance of the particles. These results were validated by numerical simulations and confirmed in posterior work.[15]

The authors of Ref. 14 put forward a hypothesis that the discovered dramatic difference between scattering properties in electric and magnetic modes is caused by difference in electromagnetic interactions between particles in different modes. It was based on an observation that magnetic dipoles as well as electric quadrupoles do not generate tangential electric fields in the array plane, and it was assumed that this means that magnetic scatterers are not interacting with each other, so that the exciting field acting on a single particle is solely the external illumination. On the other hand, the electric-dipole scatterers interact strongly and the exciting field is affected by positional disorder, which leads to resonance broadening and damping. However, from the duality principle it is known that in fact magnetic dipole particles interact via their magnetic fields exactly as strongly as electric dipoles interact via their electric fields, which means that the phenomenon discovered in Ref. 14 must have some other physical reasons.

The goal of this paper is to study the phenomenon of resonance damping and broadening theoretically and explain the strong differences in resonance broadening in different resonant modes. To this end, we analytically study the effect of positional randomness on electromagnetic behavior of grids of resonant particles which can exhibit both electric and magnetic resonant responses. We introduce a simple model which allows us to analyze the reflective, transmitting, and absorptive properties of multiresonant grids, both in the regular and amorphous states. The theory is confirmed by numerical simulations using an example of the same metasurface as that studied in Ref. 14. The results reveal the mechanisms of resonance broadening and damping in amorphous structures and explain the earlier discovered differences in the cases of electric (symmetric) and magnetic (antisymmetric) resonances. Understanding physical phenomena which define the differences between effective electromagnetic responses of regular and disordered metamaterials is urgently needed before the emerging amorphous metamaterials can find applications. Developing analytical models of disordered structures will allow the design and optimization of future composite materials with desired performance.

II. ANALYTICAL THEORY OF PLANAR ARRAYS WITH ELECTRICALLY AND MAGNETICALLY RESONANT INCLUSIONS

Let us consider an optically dense planar array of optically small resonant particles excited by normally incident plane waves. We assume that the distance between the particles in the grid a is smaller than the wavelength. We are interested in the case when each particle exhibits both electric and magnetic responses, that is, both electric and magnetic moments are induced by local electric and magnetic fields, respectively. We also assume that bi-anisotropic magnetoelectric coupling is either forbidden due to the particle symmetry or it is negligible. Many widely studied infra-red and optical metamaterial structures like the cut-wire pairs considered in Ref. 14 belong to this class. In this paper we consider only electric and magnetic dipole moments of particles, neglecting quadrupoles and higher-order moments, concentrating on the influence of array randomness on the reflection and transmission coefficients. Relative strengths of dipolar and higher-order effects in cut-wire pairs have been analyzed in Ref. 22.

Assuming for simplicity that no cross-polarized dipole moments in the array plane are induced (the particles have the form of disks or squares, for example) and considering the excitation by normally incident plane waves, we can write the relations between the induced electric dipole moment p, magnetic dipole moment m, and the incident fields $E_{\rm inc}$ and $H_{\rm inc}$ as scalar relations

$$p = \alpha_{ee}(E_{\rm inc} + \beta_{ee} p), \quad m = \alpha_{mm}(H_{\rm inc} + \beta_{mm} m). \tag{1}$$

Here α_{ee} and α_{mm} are the electric and magnetic polarizabilities of individual inclusions, respectively. Parameters β_{ee} and β_{mm} are called *interaction constants* and they measure contributions of the fields created by all other particles of the array into the local field $E_{\rm loc} = E_{\rm inc} + \beta_{ee} p$ exciting each particle (see, e.g., Ref. 23). The interaction constants for electric and magnetic dipoles are related simply as

$$\beta_{mm} = \frac{1}{\eta_0^2}\beta_{ee}, \tag{2}$$

where $\eta_0 = \sqrt{\mu_0/\varepsilon_0}$ is the wave impedance of the surrounding space. Fields created by magnetic dipoles do not contribute to the electric local field exciting electric dipoles because the tangential component of the electric field of the magnetic dipole grid equals zero in the array plane. Likewise, fields scattered by electric dipoles do not excite magnetically polarizable particles positioned in the same plane. Most often, both moments are actually induced in the same particles, but the two modes have resonances at different frequencies.

Next we calculate the plane-wave electric fields created by the surface averaged electric current sheet $J_e = -\frac{i\omega p}{a^2}$ and the magnetic current sheet $J_m = -\frac{i\omega m}{a^2}$ (the harmonic time dependence assumption is of the form $e^{-i\omega t}$ and a is the period of the grid):

$$E_{\rm ref}^{\rm e} = -\frac{\eta_0}{2}J_e, \quad H_{\rm ref}^{\rm m} = -\frac{1}{2\eta_0}J_m, \tag{3}$$

$$E_{\rm ref}^{\rm m} = -\eta_0 H_{\rm ref}^{\rm m}, \quad E_{\rm ref} = E_{\rm ref}^{\rm e} + E_{\rm ref}^{\rm m}. \tag{4}$$

Here $E_{\rm ref}^{\rm e}$ and $E_{\rm ref}^{\rm m}$ are reflected electric fields created by the induced electric and magnetic currents J_e and J_m, respectively, and $H_{\rm ref}^{\rm m}$ is the reflected magnetic field created by the induced magnetic current J_m. Solving (1) for the induced dipole moments in terms of the incident fields and using (3) and (4) we find the reflection and transmission coefficients in the simple form

$$R = \frac{E_{\rm ref}}{E_{\rm inc}} = R_e + R_m = \frac{i\omega\eta_0}{2a^2}\frac{1}{\frac{1}{\alpha_{ee}} - \beta_{ee}} - \frac{i\omega}{2\eta_0 a^2}\frac{1}{\frac{1}{\alpha_{mm}} - \beta_{mm}}, \tag{5}$$

$$T = 1 + R_e - R_m. \tag{6}$$

Note that the minus sign before the last term in (5) and (6) is due to the electric field definition of the partial reflection and transmission coefficients $R_e = E_{\rm ref}^{\rm e}/E_{\rm inc}$ and $R_m = E_{\rm ref}^{\rm m}/E_{\rm inc}$. Here we have used the plane-wave relation between the electric and magnetic incident fields: $H_{\rm inc} = E_{\rm inc}/\eta_0$. The two partial reflections coefficients R_e and R_m correspond to the fields created by the induced electric and magnetic currents, respectively. Since β_{ee} has the dimension of $1/(\varepsilon_0 a^3)$ and β_{mm} has the dimension of $1/(\mu_0 a^3)$, it is convenient to multiply and divide the reflection coefficients by $\varepsilon_0 a^3$ or $\mu_0 a^3$. The result is

$$R_e = \frac{ik_0 a}{2}\frac{1}{\frac{\varepsilon_0 a^3}{\alpha_{ee}} - \beta}, \tag{7}$$

$$R_m = -\frac{ik_0 a}{2}\frac{1}{\frac{\mu_0 a^3}{\alpha_{mm}} - \beta}, \tag{8}$$

where $k_0 = \omega\sqrt{\varepsilon_0\mu_0}$ is the wave number in the surrounding space. The normalized dimensionless interaction constants are the same for both electric and magnetic particles, and we denote them as β:

$$\beta = \varepsilon_0 a^3 \beta_{ee} = \mu_0 a^3 \beta_{mm}. \tag{9}$$

Let us assume a simple Lorentz-type resonant response model of individual particles. This type of resonant response is very common and approximates very well the particle response near their resonances. Let us write down the *inverse* values of the normalized polarizabilities to make it easy to discuss the radiation loss factor:

$$\frac{\varepsilon_0 a^3}{\alpha_{ee}} = \left(\frac{A_e}{\omega_{0e}^2 - \omega^2 - i\omega\Gamma_e}\right)^{-1} - i\frac{k_0^3 a^3}{6\pi}, \tag{10}$$

$$\frac{\mu_0 a^3}{\alpha_{mm}} = \left(\frac{A_m}{\omega_{0m}^2 - \omega^2 - i\omega\Gamma_m}\right)^{-1} - i\frac{k_0^3 a^3}{6\pi}. \tag{11}$$

Here $\Gamma_{e,m}$ model the dissipation losses in the particle (in respective modes), while the last imaginary term is due to the scattering (reradiation of power) loss.[23,24] In case of regular or "totally random" grids (on the wavelength scale) there is no scattering loss, when the array period is smaller than the wavelength. The term "totally random" means a structure which appears uniform at the scale of the wavelength. This is possible when the numbers of inclusions inside any area of the size $\lambda \times \lambda$ is large and the distances between the inclusions are small. In this case spherical-wave scattering from individual particles is suppressed by interactions between the particles in the array. Correspondingly, the imaginary parts of the interaction constants β_{ee} and β_{mm} contain terms proportional to k_0^3 which compensate the corresponding terms in the inverse polarizabilities (see, e.g., Refs. 23 and 25):

$$\beta_{\rm regular} = {\rm Re}(\beta) - i\frac{k_0^3a^3}{6\pi} + i\frac{k_0a}{2}. \tag{12}$$

The other imaginary term corresponds to the plane waves created by the surface-averaged currents. This term can be derived from the definition of the normalized interaction constant $\beta = \epsilon_0 a^3 E^{\rm e}_{\rm ref}/p$, where only the plane-wave component of the field should be retained. Substituting $E^{\rm e}_{\rm ref} = -\frac{\eta_0}{2}J_e$ and $J_e = -\frac{i\omega p}{a^2}$ we find that this component of the interaction constant indeed reads $\beta = i(k_0a)/2$. In case of amorphous (on the wavelength scale) arrays particles scatter individually, and there is no corresponding term in the interaction constants:

$$\beta_{\rm amorph} = {\rm Re}(\beta) + i\frac{k_0a}{2}. \tag{13}$$

In the quasistatic limit ${\rm Re}(\beta) \approx 0.36$ (see Ref. 23).

Next, we substitute these interaction constants and the Lorentz particle polarizabilities (10) and (11) in the general formulas for the reflection coefficients (7) and (8). For regular or totally random (on the wavelength scale) arrays we get

$$R_{e\,\rm regular} = i\frac{k_0a}{2}\frac{A_e}{\tilde\omega_{0e}^2 - \omega^2 - i\omega\Gamma_e - i\frac{k_0a}{2}A_e}, \tag{14}$$

$$R_{m\,\rm regular} = -i\frac{k_0a}{2}\frac{A_m}{\tilde\omega_{0m}^2 - \omega^2 - i\omega\Gamma_m - i\frac{k_0a}{2}A_m}. \tag{15}$$

Here $\tilde\omega_0$ denotes the resonant frequency shifted due to interactions between the particles in the grid. In the quasistatic approximation for the real part of the interaction constant

$$\tilde\omega_{0e,m}^2 \approx \omega_{0e,m}^2 - 0.36A_{e,m}. \tag{16}$$

For amorphous grids we get

$$R_{e\,\rm amorph} = i\frac{k_0a}{2}\frac{A_e}{\tilde\omega_{0e}^2 - \omega^2 - i\omega\Gamma_e - \frac{ik_0^3a^3}{6\pi}A_e - i\frac{k_0a}{2}A_e}, \tag{17}$$

$$R_{m\,\rm amorph} = -i\frac{k_0a}{2}\frac{A_m}{\tilde\omega_{0m}^2 - \omega^2 - i\omega\Gamma_m - \frac{ik_0^3a^3}{6\pi}A_m - i\frac{k_0a}{2}A_m}. \tag{18}$$

Let us consider the case when electric and magnetic resonances occur at different frequencies. Then in the vicinity of one of the resonances the nonresonant moment varies weakly with the frequency and we can find a simple estimation of the resonant curve width (on the field-strength scale):

$$2\Delta\omega_{e,m\,\rm regular} = \Gamma_{e,m} + \frac{k_0a}{2}\frac{A_m}{\tilde\omega_{0e,m}} \tag{19}$$

for regular grids and

$$2\Delta\omega_{e,m\,\rm amorph} = \Gamma_{e,m} + \frac{k_0^3a^3}{6\pi}\frac{A_{e,m}}{\tilde\omega_{0e,m}} + \frac{k_0a}{2}\frac{A_m}{\tilde\omega_{0e,m}} \tag{20}$$

for amorphous grids.

We now see that if the condition

$$\tilde\omega_{0e,m}\frac{\Gamma_{e,m}}{A_{e,m}} + \frac{k_0a}{2} \gg \frac{k_0^3a^3}{6\pi} \tag{21}$$

is satisfied, near the corresponding resonant frequency $\tilde\omega_{0e,m}$ the effect of inclusion position randomness is negligible, and the response of regular and amorphous structures is nearly the same. Physically, this condition means that absorption (the first member of the left-hand side) and coherent plane-wave reflection (the second member on the left) dominate over scattering (the right-hand side term). The above relation shows that this is the case of high dissipative losses, low resonance strength, and small electrical size of the unit cell. Note that for the case of negligible absorption, this condition simply tells that scattering loss is negligible in random arrays if the distance between particles is optically very small ($k_0^2a^2 \ll 3\pi$).

From the above results we can conclude that the effect of strong widening of the resonant curve of the electric-dipole mode and hardly any effect of array randomness on the magnetic mode discovered in Ref. 14 can be due to two reasons:

(1) At the frequency of the magnetic resonance the grid is practically homogeneous on the wavelength scale ("totally random"). Then the scattering term cancels out just like for periodical grids, and there is no difference in the resonant curve widths for regular and amorphous layers.

(2) At the magnetic resonance the particles are considerably more lossy and weaker excited than at the electric resonance, that is, (21) is satisfied near the magnetic resonance but not satisfied near the electric-mode resonance.

III. EXAMPLE: ARRAYS OF CUT-WIRE PAIRS

As an example we consider the cut-wire pair structure which was studied in Refs. 14, 26, and 27. A unit cell of the infinite regular array is depicted in Fig. 1. The dimensions are the same as in Ref. 14. The square lattice has the period of $a =$ 512 nm along two transverse directions. The width of the cut-wire pairs in both lateral directions is $W_c = 180$ nm. The height of each gold pair is $H_c = 30$ nm. The gap between the two elements in each pair equals $g = 45$ nm and it is filled with a material with the relative permittivity equal to $\varepsilon_{r1} =$ 1.72. The structure is placed on top of a substrate with the permittivity of $\varepsilon_{r2} = 1.5$. The permittivity of gold is taken from Ref. 28.

First we calculate the reflection and transmission coefficients for the regular array using the full-wave numerical simulator Ansoft HFSS. Substituting the numerical data for reflection and transmission coefficients in (7) and (8) and using the quasistatic approximation for the real part of the interaction constants $\beta \approx 0.36$, we extract the polarizabilities of individual inclusions. The results are shown in Fig. 2, and it is apparent that the array has electric and magnetic resonances in different frequency regions, as expected. In fact the array is weakly bi-anisotropic due to the presence of the substrate (omega-type magnetoelectric coupling[29,30]), which has been neglected in the theory and in the parameter extraction. We have checked that this approximation is valid by repeating the simulations and parameter extraction for the same array in free

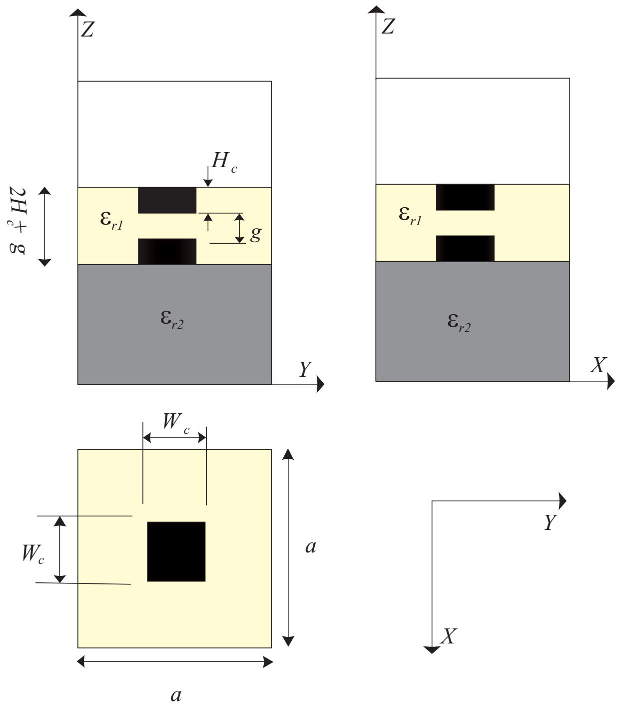


FIG. 1. (Color online) Geometry of the unit cell of the cut-wire array.

space. The results are presented in Fig. 3 and they show that this simplifying assumption is reasonable: The substrate effect is quite small. Behavior of the extracted electric and magnetic polarizabilities is very close to the canonical Lorentz-type resonant response.

Scattering losses which appear in transition from regular to amorphous grids we model by the randomness parameter $0 \leqslant r_n \leqslant 1$, where unity corresponds to the case where the scattering loss is completely compensated *(regular array)* and $r_n = 0$ means that the scattering loss is not compensated at all *(amorphous array, each inclusion scatters individually)*. Transition from regular to amorphous state we model modifying the interaction constant (12) as follows:

$$\beta = \mathrm{Re}\{\beta\} - r_n \frac{i k_0^3 a^3}{6\pi} + \frac{i k_0 a}{2}, \tag{22}$$

which corresponds to a continuous transition from (12) to (13) with r_n changing from unity to zero. It should be noted that for simplicity the randomness factor r_n is assumed to be the same for both electric and magnetic interaction constants. Due to differences in resonant frequencies, this means that the same value of r_n may correspond to somewhat different degrees of geometrical randomness of particle positions.

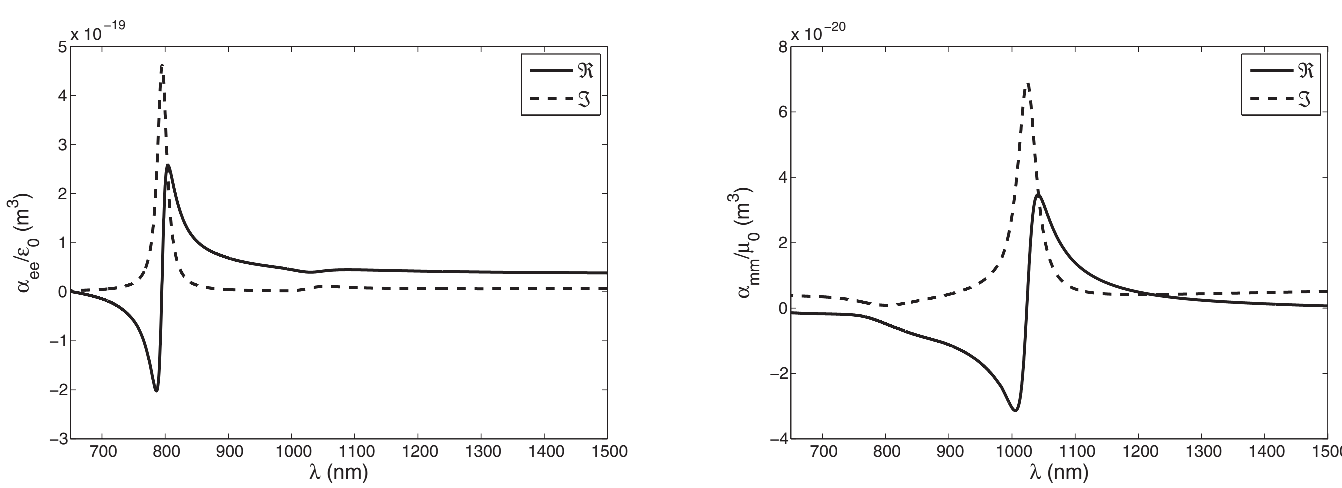
It is clear that in the general case one has to modify also the real part of interaction constant ($\mathrm{Re}\{\beta\}$) in order to fully consider the effect of randomness on the resonance broadening and damping. The real part of the interaction constant measures the reactive part of the interaction field (near-field coupling with the neighboring inclusions), and its random fluctuations lead to random shifts of the resonant frequencies of the particles and broadening of the resonant curve of the random array. The very good agreement of our simple model (which ignores this effect) with the experiment indicates that the effect of this random frequency shift is in the studied case negligible. Let us estimate these random variations of the resonant frequency. The frequency shift due to particle interactions is given by (16), which we write in terms of the relative shift:

$$\frac{\tilde{\omega}_{0e} - \omega_{0e}}{\omega_{0e}} \approx -\mathrm{Re}\{\beta\} \frac{A_e}{2\omega_{0e}^2}. \tag{23}$$

The resonance strength factor A_e can be estimated from the known particle polarizability. Expecting formula (10) in the limit of small frequencies we find that

$$\frac{A_e}{\omega_{0e}^2} = \left. \frac{\alpha_{ee}}{\epsilon_0 a^3} \right|_{\omega \to 0}. \tag{24}$$

This result shows that the frequency shift is determined by the quasistatic polarizability and the array period: As expected, the effect is stronger for high polarizabilities and dense arrays. An important observation is that the polarizability value here is not



(a) (b)

FIG. 2. (a) Electric polarizability of a single cut-wire pair and (b) magnetic polarizability of the same particle.

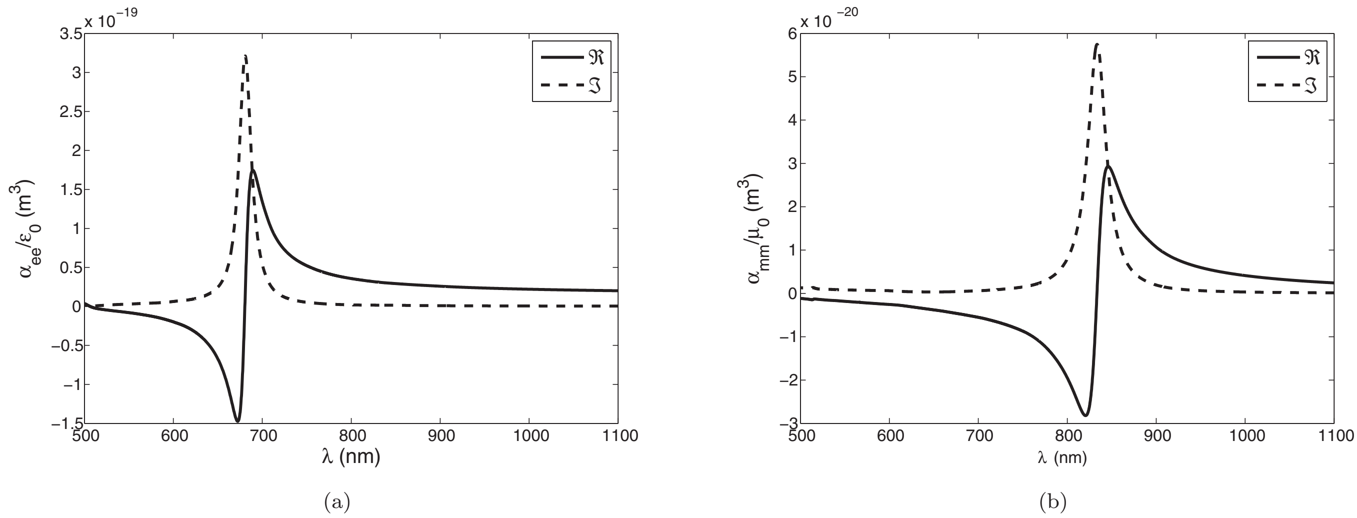


FIG. 3. (a) Electric polarizability of a single cut-wire pair in free space and (b) magnetic polarizability of the same particle.

the resonant value but the (small) low-frequency value. Now we are ready to estimate the effect for our particular example. The low-frequency value of the normalized polarizability we find from Fig. 2(a). Substituting that and $a = 512$ nm, the

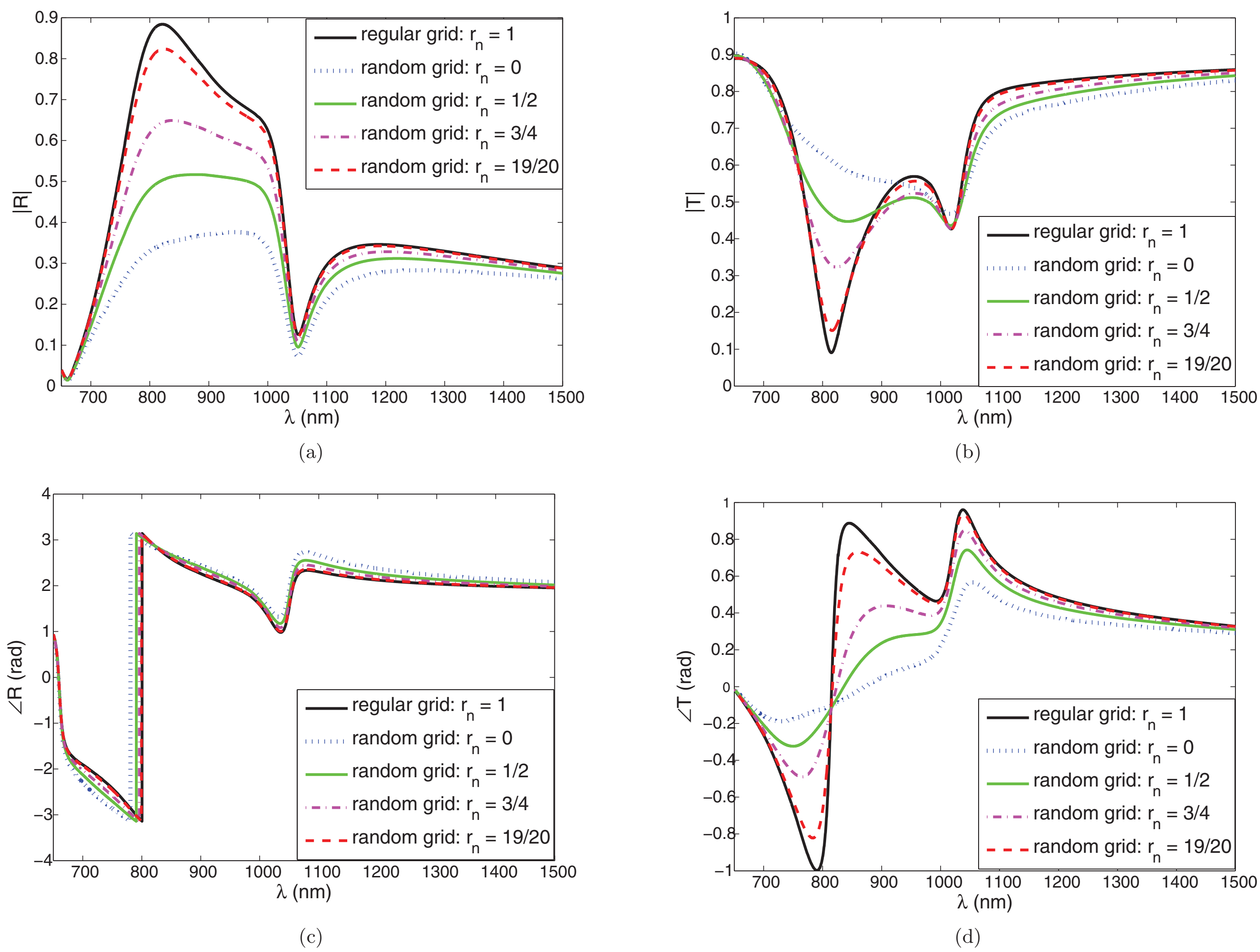


FIG. 4. (Color online) (a) Amplitude of the reflection coefficient. (b) Amplitude of the transmission coefficient. (c) Phase of the reflection coefficient. (d) Phase of the transmission coefficient for grids with different randomness levels (r_n).

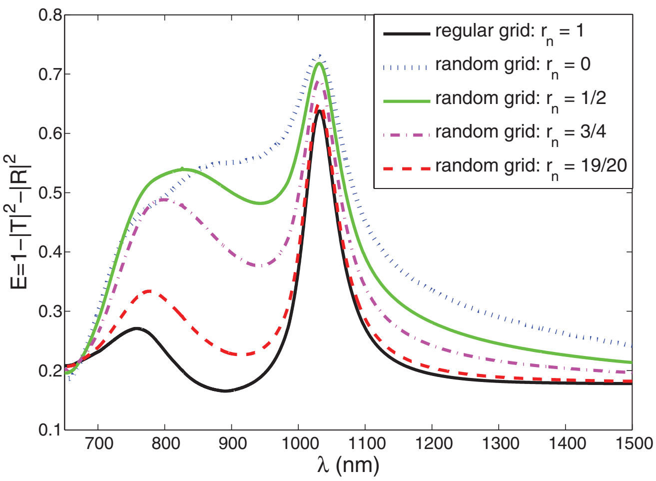


FIG. 5. (Color online) Absorption in the array in transition from regular to amorphous states.

result is

$$\frac{\tilde{\omega}_{0e} - \omega_{0e}}{\omega_{0e}} \approx -0.14\mathrm{Re}\{\beta\}. \qquad (25)$$

For the regular grid Re$\{\beta\}$ approximately equals 0.36. Assuming that in random arrays it may have 50 percent deviation from this value, we come to an estimation of this frequency shift in the range of 0.02–0.07, which is clearly negligible. The case of magnetic mode can be considered analogously, but in that case we cannot use the low-frequency limit of (11) to estimate A_m, because in that limit A_m vanishes. However, from duality we expect that the magnetic-mode frequency shift will be of the same order as for the electric mode.

Next we investigate how the reflection, transmission, and extinction change in transition from regular to amorphous states, using the analytical formulas (7) and (8) with the extracted values of the polarizabilities and the interaction constant (22). Figures 4 and 5 show the randomness effects. One can see that the developed simple model gives very good agreement with the experimental and numerical data from Ref. 14. Electrical response of the grid is strongly influenced by randomness, while close to the magnetic resonance there is almost no dependence on randomness. The reason for this phenomenon is the difference in the ratio of the absorption and scattering losses. Figure 5 shows that in the periodical case the absorption is much stronger at the magnetic resonance than at the electric one. Higher losses are mainly due to larger imaginary part of gold permittivity, which is more than two times higher at the magnetic resonance: $\mathrm{Im}(\varepsilon)_{1022\,\mathrm{nm}} \approx 3.2$, $\mathrm{Im}(\varepsilon)_{797\,\mathrm{nm}} \approx 1.5$.

In addition, for the case of the grid in free space (Fig. 3) we have fitted the numerically extracted polarizability curves to the Lorentz model (10) and (11) and extracted parameters $\Gamma_{e,m}$ and $A_{e,m}$. This allowed us to find the values in inequality (21). At the resonant frequency of the electric polarizability we find that the left-hand side equals $0.4 + 2.3$, while the right-hand side equals 5.6. Scattering effects are clearly dominating and position randomness changes the array response quite significantly. At the resonant frequency of the magnetic polarizability the left-hand side reads $2.3 + 2$, while the right-hand side equals 3. In this case the terms are of the same order and the randomness effect is much weaker. Note that condition (21) is a simple approximation which assumes that the two resonances are sharp and well separated. In this particular example, in the frequency region of the magnetic resonance the electric dipoles in fact give a significant contribution to the total absorption and coherent reflection.

IV. CONCLUSIONS

In this paper we have developed a simple model which explains the electromagnetic effects in transition from regular to random states of resonant particle arrays. We have derived a general condition under which randomizing particle positions gives only negligible effects on the reflection and transmission coefficients and explained the earlier discovered dramatic differences in resonance damping for electric and magnetic modes of particles. We have also shown that the physical phenomena leading to the resonance damping in amorphous structures are the same for electrically or magnetically polarizable particles. The widening of the resonances takes place due to additional scattering losses, which are compensated in the case of electrically dense periodical grids.

Studying transition to the amorphous state for a particular example of cut-wire pairs we have found that the reason for the much weaker resonance widening and damping in the magnetic mode is strong absorption in that frequency range. When scattering losses are much smaller than the dissipation losses, they make little impact on the total extinction. On the contrary, at the higher-frequency electric resonance scattering losses are stronger than the dissipation ones, which leads to strong resonance damping and distortion in the random case. In other situations, different transition effects in different resonant modes can also be caused by differences in the electrical size of the unit cell. In the considered example, the array period is comparable with the wavelength, thus, even for geometrically random positions of the particles with respect to the cell centers, the array cannot be made homogeneous on the wavelength scale.

Our findings can have important implications in understanding the physical differences in electromagnetic responses of regular and amorphous structures, in design of various metamaterial structures for such applications as subwavelength imaging, control of thermal radiation, microwave, terahertz and optical absorbers, and others. Using the developed model it is possible to predict and engineer the effects of randomness, relaxing conventional requirements on strong periodicity and make use of inexpensive self-assembly techniques in production of metamaterials.

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