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Strong terahertz absorption bands in a scaled plasmonic crystal

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The authors show the existence of strong absorption bands at terahertz frequencies in scaled plasmonic crystals composed of a periodic arrangement of two-dimensional electron-gas shells. The absorption bands appear as a result of the excitation of plasmon modes supported by the shells and can be tuned throughout the entire terahertz spectral region by varying the electron-gas density in the shells. These plasmonic crystals exhibit promising properties enabling their applications as tunable terahertz filters, absorbers, and thermally activated sources. © 2007 American Institute of Physics. [DOI: 10.1063/1.2749869]

Photonic crystals have been extensively studied for nearly three decades^{1,2} and recent progress in nanopatterning has allowed achieving full photonic band gaps in the visible and near-infrared (NIR) spectral regions,³ mainly arising from strong Bragg scattering. The polaritonic interaction between photons and intrinsic eigenmodes localized in the unit cells of a crystal can also lead to robust band gaps. This type of situation is encountered, for instance when considering resonant atoms,⁴ localized plasmons,⁵ or excitons.⁶ In the terahertz domain, the Bragg scattering cannot play a role in the electromagnetic response of nanoscale photonic crystals, since the dimension of the nanocrystal unit cell is much less than the wavelength of the terahertz radiation. However, a localized intrinsic resonance might significantly affect the terahertz response of the nanocrystal if the frequency of this resonance falls into the terahertz spectral region. Similar to visible and NIR plasmon resonances in metals, low-dimensional semiconductor nanostructures exhibit plasmon resonances at terahertz frequencies, thus becoming natural candidates to realize the terahertz plasmonic crystal.

While the plasmons in planar two-dimensional (2D) electron systems have been broadly investigated, a live interest to the electron kinetics in curvilinear quantum wells has arisen quite recently.⁷⁻⁹ Curvilinear quantum wells can be produced by different methods.^{8,10-13} In Refs. 8 and 13 using the pyroelectric-semiconductor structures was suggested. These structures can be composed of either the pyroelectric spherical inclusions ingrained into a semiconductor matrix or, as the inverse structure, spherical semiconductor inclusions ingrained into a pyroelectric matrix. As a result of the spontaneous polarization inherent in pyroelectric material, spherical semishells of 2D electron gas can be formed at the inclusion boundaries. Such 2D electron-gas shells, when ar-

ranged in a three-dimensional (3D) periodical lattice, form a scaled plasmonic crystal that should exhibit the complex electromagnetic response associated with collective plasmon excitations.

In this letter, we report on broad plasmonic bands of strong terahertz absorption that are formed in a scaled plasmonic crystal composed of 2D electron-gas open shells. The nature and symmetry of the shell modes are discussed. Changing the 2D electron density in the shells is shown to move plasmon absorption bands throughout the entire terahertz spectral domain.

In order to elucidate how plasmon excitations behave in scaled plasmonic crystals, let us first consider a single spherical thin electron-gas shell. It is well known that a conductive sphere can support Mie plasmon modes.¹⁴ In a conductive shell, each of these modes splits into a doublet of spherelike and voidlike modes with in-phase and out-of-phase charge oscillations at opposite (outer and inner) surfaces of the shell wall, respectively.^{15,16} We consider small shells where magnetic multipoles can be safely neglected, so that the mode frequencies are given by the poles of the electric polarizability.¹⁴ In the quasielectrostatic limit, this yields the following equation for the plasmon eigen-frequencies:

$$\frac{l\varepsilon_1 + l\varepsilon_2(\omega) + \varepsilon_2(\omega)}{\varepsilon_1 - \varepsilon_2(\omega)} = fl(l+1) \frac{\varepsilon_3 - \varepsilon_2(\omega)}{l\varepsilon_2(\omega) + l\varepsilon_3 + \varepsilon_3}, \quad (1)$$

where l is the orbital momentum quantum number, ε_1 and ε_3 are the dielectric constants of materials in the core and outside the shell, respectively, $\varepsilon_2(\omega)$ is the frequency-dependent dielectric function of the material forming the shell, $f = a^{2l+1}/(a+h)^{2l+1}$, a is the shell inner radius, and h is the shell-wall thickness. In particular, the dipolar modes ($l=1$) are known as the Fröhlich modes.¹⁴ We describe the electromagnetic response of the electrons inside the shell wall using the local Drude model,

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FIG. 1. (Color online) Terahertz absorption cross section σ vs frequency for 2D spherical shells with areal electron-gas density $N_s = 2.5 \times 10^{12} \text{ cm}^{-2}$, inner radius $a = 45 \text{ nm}$, shell-wall thickness $h = 5 \text{ nm}$, and different angular openings (see inset). The frequencies of the spherelike (left) and voidlike (right) multipole modes of a full shell are marked by arrows, as calculated in the quasielectrostatic limit for $l = 1, 2$, and 3 . The cross section is normalized to the projected area of the shell, $\pi(a+h)^2$. The polarization of the electric field E_{in} in the incident terahertz radiation is shown in the inset.

$$\varepsilon_2(\omega) = \varepsilon_b \left(1 - \frac{\omega_p^2}{\omega(\omega + i\nu)} \right), \quad (2)$$

where $\omega_p = \sqrt{4\pi e^2 N_s / (\varepsilon_b m^* h)}$ is the bulk plasmon frequency, N_s is the areal electron density in the shell wall, e and m^* are the electron charge and effective mass, respectively, and ν is a phenomenological free electron scattering rate. If $\nu = 0$ and $\varepsilon_1 = \varepsilon_3 = \varepsilon_a$, the frequencies of Fröhlich resonances are given by

$$\omega_{\pm}^2 = \frac{4\varepsilon_p^2(1-f)}{4\varepsilon_b(1-f) + \varepsilon_a(5+4f \mp 3\sqrt{1+8f})}, \quad (3)$$

where the lower and upper signs refer to spherelike and voidlike modes, respectively. For a 2D electron shell with $N_s = 2.5 \times 10^{12} \text{ cm}^{-2}$, $m^* = 0.242m_e$ (m_e is the free electron mass), $\varepsilon_a = \varepsilon_b = 9$, inner shell radius $a = 45 \text{ nm}$, and shell-wall thickness $h = 5 \text{ nm}$, the frequencies of spherelike and voidlike plasmons are $\omega_- = 3.43 \text{ THz}$ and $\omega_+ = 13.1 \text{ THz}$, respectively. These parameters are characteristic for granular

pyroelectric/semiconductor systems based on GaN.⁸

Figure 1 shows the terahertz absorption cross section of a shell with different angular openings, calculated in the framework of a rigorous electromagnetic boundary element method¹⁷ (BEM) and assuming $\nu = 4.4 \times 10^{13} \text{ s}^{-1}$ in Eq. (2) corresponding to the electron mobility in GaN of $1650 \text{ cm}^2/\text{V s}$. Using the BEM, the exact solution of Maxwell's equations in the frequency domain is reduced to a set of self-consistent integral equations for the field boundary conditions, written in terms of auxiliary surface charges and currents that are solved through linear algebra upon discretization of the integrals. In the spectra of a full shell the resonances are exhibited at frequencies that are in good agreement with the frequencies of spherelike and voidlike Fröhlich modes calculated above in the quasielectrostatic limit for $\nu = 0$. It follows from Fig. 1 that these two different (spherelike and voidlike) modes resonate also in open (even in shallow) 2D electron shells. However, the intensity of the resonances is weaker in shallow shells due to their comparatively small volume. Besides, the dipole spherelike mode is flanked by a sizable quadrupolelike mode ($l=2$) in open shells.

In order to assess the difference between spherelike and voidlike modes, we have calculated the electric field distribution of these resonances at the same moment of the optical cycle (see Fig. 2). For a better understanding, we also present schematically in Figs. 2(a) and 2(b) the electric field lines and oscillating charge distributions for the full shell. These results demonstrate that the net dipole moment of the voidlike mode is smaller than that of the spherelike mode, which explains weaker resonances of the voidlike plasmons in Fig. 1 compared to the spherelike modes. Figure 2(b) clearly shows that the field of the voidlike mode of a full shell is localized inside the shell wall, since charges of opposite sign are induced on either side of the wall in this mode. In contrast, the electric field of the spherelike mode [see Fig. 2(a)] concentrates in the shell core with charges of the equal sign induced on both sides of the shell wall. In the dipole mode shown in Figs. 2(a) and 2(b), the electric field of both spherelike and voidlike modes is homogeneous inside the full-shell core. Figures 2(c) and 2(d) show the electric field distributions in spherelike and voidlike modes of an open shell with angular opening $\theta = 130^\circ$ (see inset in Fig. 1). Similarly

FIG. 2. (Color online) Calculated electric-field distributions at the resonances of [(a) and (c)] spherelike and [(b) and (d)] voidlike modes of [(a) and (b)] a full shell and [(c) and (d)] an open shell with angular opening $\theta = 130^\circ$ at the same moment of the optical cycle. The magnitude of the electric field amplitude is normalized to the magnitude of the electric field in the incident terahertz radiation. The other parameters are the same as in Fig. 1. Electric field lines and charge distributions for each plasmon mode of a full shell are shown schematically in panels (a) and (b).

FIG. 3. Terahertz absorption spectra of a plasmonic crystal slab composed of 130 [111] layers of 2D electron shells with $a=45 \text{ nm}$ and $h=5 \text{ nm}$ arranged in a fcc lattice for (a) the full shells and (b) open shells with angular opening $\theta=130^\circ$ with different values of the areal electron density: $N_s=10^{12} \text{ cm}^{-2}$ (solid curves), $2.5 \times 10^{12} \text{ cm}^{-2}$ (dashed curves), and $5 \times 10^{12} \text{ cm}^{-2}$ (dotted curves).

to the full shell, the field of the voidlike mode concentrates inside the shell wall, while the spherelike mode spreads out from the shell and is strongly enhanced at the edges ($E/E_{\text{in}} \approx 26$), due to the buildup of oscillating charges.

Now, we will turn our attention to plasmonic crystals formed by shells arranged into the 3D lattice. Figure 3 shows the terahertz absorption spectra of multilayered plasmonic crystal slabs composed of 130 [111] layers of shells ordered in a face-centered-cubic (fcc) lattice (the [111] plane of the lattice is parallel to the slab surface), both for full shells [Fig. 3(a)] and for open shells with angular opening $\theta=130^\circ$ [Fig. 3(b)]. The fcc lattice constant is $d=212 \text{ nm}$. The optical spectra of the plasmonic crystals have been calculated using a self-consistent electromagnetic multiple-scattering layer-Korringa-Kohn-Rostoker approach,¹⁸ into which we have introduced scattering matrices of open shells obtained from the BEM.¹⁷ Strong terahertz absorption bands emerge in the vicinity of spherelike and voidlike plasmon resonance frequency. This corresponds to the presence of plasmonic bands of the crystal. The plasmonic absorption bands considered here are radically different from conventional photonic band gaps of a photonic crystal, where photon transmission is forbidden. While a single full shell does not exhibit higher-order multipolar plasmon resonances beyond dipolar one due to its perfect spherical symmetry, the sizable contribution of quadrupolar components appears in the fine structure of the low-frequency absorption band when shells are arranged into the lattice [see Fig. 3(a)]. Figure 3 clearly demonstrates that the absorption bands can be tuned in frequency throughout the entire terahertz spectrum by changing the 2D electron density in shells within a reasonable range of values, which might be accessible to external control.

In conclusion, we have demonstrated the existence of the absorption bands in a scaled plasmonic crystal at terahertz frequencies. These absorption bands appear as a result of plasmon mode excitation in (open) shells of the crystal and can be tuned in frequency throughout the entire terahertz domain by changing the areal electron density in the shell, which can be realized in turn by applying external electric and/or magnetic fields or through light irradiation of the plas-

monic crystal slab. Such plasmonic crystals may find various applications as tunable terahertz filters, absorbers, and thermally activated sources.

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