Monolithic metallic nanocavities for strong light-matter interaction to quantum-well intersubband excitations

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Abstract: We present the design, realization and characterization of strong coupling between an intersubband transition and a monolithic metamaterial nanocavity in the mid-infrared spectral range. We use a ground plane in conjunction with a planar metamaterial resonator for full three-dimensional confinement of the optical mode. This reduces the mode volume by a factor of 1.9 compared to a conventional metamaterial resonator while maintaining the same Rabi frequency. The conductive ground plane is implemented using a highly doped n+ layer which allows us to integrate it monolithically into the device and simplify fabrication.

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References and links

1. Introduction

Light-matter interaction is a fascinating and highly active field of research [1]. In a simplified picture, a two-level system is coupled to an optical cavity which causes both systems to interact and therefore exchange energy. Depending on the efficiency of this energy exchange...
in relation to all the loss mechanisms present in the system, e.g. non-radiative decay of the two-level system or light escaping the cavity (optical loss), different physical regimes can be studied. In the weak coupling regime, the spontaneous lifetime of the two-level system can be reduced through the so-called Purcell effect [2, 3]. Here we will focus on the strong coupling regime, where the energy exchange between the dipole and the optical cavity is faster than all loss mechanisms combined. Energy oscillates between the cavity and the two-level system multiple times before it is lost. The characteristic signature of this energy transfer is a beating in the time domain of different optical signals (observable in emission, reflection or absorption) or a splitting of the single cavity resonance into two polariton branches in the frequency domain. This process happens at a characteristic rate called the vacuum Rabi frequency $\Omega_R$. Typically the Rabi frequency is only a very small fraction of the fundamental optical carrier frequency. In other words, the optical dipole undergoes many optical cycles before it exchanges energy with the cavity. To observe this splitting experimentally it is therefore necessary to use cavities with a high quality factor. This makes low-loss dielectric resonators the only feasible choice at optical wavelengths [4].

Significantly larger polariton splittings compared to the bare cavity resonance frequency have been demonstrated recently [5–8]. These experiments used optical dipoles with extremely large dipole matrix elements to achieve strong interaction; typical examples are organic molecules (J-aggregates) [5, 6] or intersubband transitions (ISTs) in semiconductor quantum-wells (QWs) [7, 8]. The resulting large splittings (values beyond 50% of the carrier frequency have been reported [8]) lower the requirement for the optical cavity and make the experimental observation of strong coupling signatures much easier. Therefore, a larger variety of potential structures can now be used as optical cavities including metallic structures such as gratings [9] or frequency selective surfaces [10]. Their main advantage is an extreme spatial confinement of the optical mode to deep-subwavelength volumes based on localized surface plasmons [11, 12]. Furthermore, the large enhancements in the electric field close to resonant metallic surfaces support the strong light-matter interaction even further.

Recently, metallic metamaterial resonators have been used for studying light-matter interaction and particularly in the strong coupling regime [8, 10, 13, 14]. These resonators are of subwavelength dimensions and can be designed to support electric or magnetic resonances. Planar arrays of these resonators have been recently used in a number of applications such as sensing [15], modulation of light [16] and beam control [17–21]. Planar metamaterial resonators can be straightforwardly combined with optical dipoles implemented using optical transitions in semiconductor QWs. Thin layers of semiconductors with different bandgaps form potential wells for carriers that inhibit the free electron motion along one spatial direction. Electrons present in these QWs can only occupy discrete energy levels resulting in atom-like absorption and emission spectra when they are promoted between these energy levels. The optical properties such as transition energy or dipole matrix element of these ISTs are controlled by the thickness of the individual wells and barriers, and become largely independent of the underlying semiconductor material. One consequence of the quantization of energy levels along only one spatial dimension that is essential to this work is the anisotropy of the dielectric permittivity. Only light that is polarized along the growth direction (in this manuscript referred to as z-axis) couples to the ISTs due to the dipole selection rules [22]. Therefore, normal incidence radiation (which propagates along the z direction and will be used in this manuscript to study the strong light-matter interaction) will not be able to couple to ISTs. Metamaterial resonators can be used to solve this coupling problem. In the near-field region of the resonators, most of the incoming optical field is transferred to the perpendicular field component at the metamaterial resonance [14]. This makes planar metamaterials an excellent platform to study light-matter interaction in sub-wavelength volumes.
For an even stronger reduction in interaction volume it is necessary to study the cavity near-field in more detail. Designing and controlling the spatial extent of the optical mode in the lateral direction is fairly straightforward; the mode is restricted predominantly to the regions directly underneath the metal traces. However, in the vertical direction (perpendicular to the metamaterial) the electric field extent is given by the decay length of the cavity mode. This decay length is difficult to control and in the mid-infrared (MIR) spectral range it is typically on the order of a few hundred nanometers [23, 24]. This vertical field profile is crucial for optimizing the position and the thickness of the QW stack; only the fraction of the cavity mode that has a spatial overlap with the QWs contributes to the strong coupling.

Here we present an approach to realizing full three dimensional mode confinement using **monolithic metamaterial nanocavities** that remains compatible with planar fabrication processes, and discuss the strong light-matter interaction using these nanocavities. The basic geometry for the device is shown in Fig. 1. We use a planar metamaterial resonator fabricated on top of a semiconductor substrate and add a bottom conductive ground plane. We realize the ground plane using a thin, highly doped n+ layer which is integrated monolithically into the device and allows for simple fabrication. The quantum well stack containing the optical dipoles can be very thin as the conducting ground plane limits the vertical extent of the optical field. This additional confinement reduces the optical cavity volume by a factor of 1.9 as compared to the use of just a metamaterial resonator with no backplane. Incorporating our semiconductor based ground plane into the device we achieve the same interaction strength between the ISTs and the cavity field (i.e., the same Rabi frequency) using one-third the QW stack thickness of a comparable design without ground plane.
Fig. 2. Schematic of both sample geometries. The In$_{0.53}$Ga$_{0.47}$As/Al$_{0.48}$In$_{0.52}$As quantum-wells are designed for a transition energy of 100 meV (top left). Both samples are grown on an InP-substrate (blue); the quantum-well stack is indicated in purple. The monolithic metamaterial nanocavity sample (bottom right) is confined by an 800 nm thick n' layer (red) in the vertical direction. The permittivity of the n' layer is shown in the bottom left subplot where $\varepsilon'$ refers to the real part and $\varepsilon''$ to the imaginary one. The difference in “dogbone” size between the conventional sample and the monolithic metamaterial nanocavity is required by the difference in permittivity of the QW-stack and the ground plane. We have adjusted the sizes to keep the metamaterial resonance frequency constant.

2. Metamaterial design and interaction volume

Conductive ground planes are a well-studied approach for the design of radio-frequency antennas. From a modeling standpoint we can replace the region below the ground plane with the mirror charge from above; the field profile remains identical in the entire upper half-space [25]. This approach is commonly used to reduce the length of a dipole antenna by a factor of two [26]. A monopole antenna placed perpendicularly on top of a ground plane gives the same emission profile. More recently, dipoles that are oriented parallel to the ground plane have been used to realize ultra-broadband antennas [27, 28] or small antennas based on split-ring resonators [29]. Here we will focus on another aspect when using ground planes: the efficient confinement of radiation to small volumes [30].

Instead of the antennas mentioned before, we combine the conductive ground plane with a “dogbone”-metamaterial nanocavity [31]. We chose this particular metamaterial resonator because of its strong near-field enhancement along the entire area of the resonator. We study two cases: a) a “dogbone” resonator fabricated on top of a 650 nm thick stack of QWs as the reference sample, and b) a “dogbone” resonator fabricated on top of a 195 nm thick QW-stack with the semiconductor-based ground plane (we will call this the “monolithic metamaterial nanocavity” in this work). A schematic of both samples is presented in Fig. 2. For the monolithic metamaterial nanocavity we added an 800 nm In$_{0.53}$Ga$_{0.47}$As layer underneath the thin QW-stack n-doped to a concentration of $10^{19}$ cm$^{-3}$. Its behavior can be described using a simple Drude model as indicated in Fig. 2. Below its plasma frequency of 39.3 THz the real
part of the permittivity remains negative and it shows metallic behavior [32]. Our QWs are based on an In_{53}Ga_{47}As/Al_{48}In_{52}As heterostructure which is lattice matched to the InP substrate. The QW-width is set to 12.5 nm with 20 nm barriers in between and this leads to a transition energy for the bound electrons of 100 meV (24.3 THz). Each well is doped to an electron sheet density of 1.25 × 10^{12} cm^{-2}. This basic sequence is repeated multiple times to achieve the required thickness for the QW-stack of 195 nm for the monolithic metamaterial nanocavity and 650 nm for the conventional sample. The QW-stacks are capped with a 30 nm thin Al_{48}In_{52}As layer. The monolithic metamaterial nanocavity has an additional 40 nm Al_{48}In_{52}As barrier between the QW-stack and the n+ layer. Both heterostructures are grown using molecular beam epitaxy which allows us to control the thickness of the individual layers (QWs, ground plane, buffer layers) precisely and integrate all layers monolithically into the sample. Furthermore, we analyzed both samples using x-ray diffraction measurements to ensure their crystal quality.

We study the strong light-matter interaction in the frequency domain analyzing the theoretical and experimental optical reflectance spectra obtained at normal incidence. We fabricated several arrays of metamaterial resonators by geometrically scaling the resonator and unit-cell dimensions which shifts the resonance frequency and allows us to sweep this transition across the IST. Due to the strong light-matter interaction we expect a distinct anti-crossing behavior rather than just two discrete resonances crossing each other. To understand our system we perform first a series of finite-difference time-domain (FDTD) simulations [33, 34]. We design one unit cell according to the schematics in Fig. 2 and apply periodic boundary conditions. It should be noted here that the conventional metamaterial and the monolithic nanocavity sample show the same periodicity in the xy-plane. Thereby, we can exclude any additional effect of the resonator spacing or nearest neighbor interaction on the light-matter coupling between the two different samples and focus on the interaction at the single resonator level. The parameters for the gold and the n+ layer (including doping concentration and mobility) are extracted from spectral ellipsometry measurements. The QWs are modeled as anisotropic, harmonic oscillators following the intersubband selection rules; only light polarized along the z-axis can couple to these transitions. The optical susceptibility is calculated following the procedure in [14, 23]. The center frequency of the absorption is set to 24.3 THz, and the full-width at half-maximum to 2.4 THz. Both values are confirmed by waveguide measurements using the same wafer as the conventional sample. It should be stressed here that despite the multiple scattering mechanisms affecting the spectral width of the IST, one phenomenological broadening term in the model is enough to capture the underlying behavior.

The importance of adding an additional confinement dimension to the optical fields can be seen in the spectrally and spatially resolved mode profiles shown in Fig. 3(a). The n+ layer prevents the mode from penetrating into the substrate and increases the optical or mode confinement. The mode volume is reduced from 2.49 × 10^{-3} \lambda_{\text{eff}}^3 for the conventional sample to 1.34 × 10^{-3} \lambda_{\text{eff}}^3 for the monolithic metamaterial nanocavity, where \lambda_{\text{eff}} describes the wavelength inside the semiconductor (\lambda/n). In both cases we define the mode volume following the Purcell definition for dispersive media [11, 12, 35, 36] and limit the integration volume to the semiconductor. Thereby, we can avoid the extreme field enhancement at the metal surface that leads to numerical problems and as a consequence to unphysical solutions.
Fig. 3. Field profiles and polariton branches simulated using finite-difference time-domain calculations. (a) Frequency and spatially resolved $E_z$ profiles for both samples without quantum-well interaction. The electric field is concentrated at the air-semiconductor interface and penetrates 600 nm into the conventional sample (left panel). The n" layer limits the cavity mode to the quantum-well region in case of the monolithic metamaterial nanocavity (right panel). The quantum-wells are sandwiched between a 30 nm (above, both samples) and a 40 nm (below, only metamaterial nanocavity) Al$_{0.48}$In$_{0.52}$As buffer layer which is indicated by the white, dashed lines. The absolute value of the $E_z$ component is integrated across the entire unit cell in the $xy$-plane to create both profiles. The field values are referenced against the maximum for both cases. (b) Simulated reflectance curves for both types of samples using finite-difference time-domain calculations. As the QW-stack thickness is increased for the conventional sample the splitting between the two polariton branches increases as well. The eigenfrequencies appear as maxima in reflectance in this case. The metamaterial nanocavity sample experiences the same polariton splitting as the three-time thicker conventional sample appearing as minima in reflectance. The black dashed lines represent the predicted polariton eigenfrequencies using the coupled oscillator model.

The effect of the decay length of the electric field for the resonator without the ground plane is analyzed by calculating the normal incidence reflectance spectra for three different
As illustrated in Fig. 3(b) the splitting between the two polariton branches increases with an increasing QW-stack thickness. To quantify this increased light-matter coupling we calculate the Rabi frequency using a coupled harmonic oscillator model [37] that allows us to fit the entire lineshape of the reflectance spectra for each metamaterial geometric scaling factor. The three samples with different QW-stacks show Rabi frequencies of 1.65 (50 nm QW stack), 2.34 (200 nm QW stack) and 2.64 THz (650 nm QW stack). Adding the ground plane allows us to shrink the QW-stack thickness and keep a similar Rabi splitting, as shown in Fig. 3(b). The calculated Rabi frequency for this case is 2.7 THz which is almost the same value obtained for the 650 nm thick conventional QW-stack sample. Therefore, the predicted polariton eigenfrequencies using the coupled oscillator model are also almost identical for the monolithic nanocavity and the three-time thicker QW-stack using a conventional metamaterial with no ground plane (Fig. 3(b)). The reason for the same light-matter interaction strength despite the much thinner QW-stack is the additional mode confinement provided by the ground plane.

3. Measurement results and discussion

Experimentally, we confirm our theoretical findings by measuring the normal incidence reflectance spectra for the conventional sample (650 nm QW stack) and the monolithic metamaterial nanocavity sample. All optical measurements are performed at room
temperature using a Nicolet Magna 860 Fourier-transform infrared spectrometer with a microscope objective. The “dogbone”-nanocavity is defined using electron beam lithography; a Ti/Au (5/100 nm) layer is evaporated followed by a standard lift-off process. Scanning electron micrograph images for a processed device are presented in Fig. 4(a). It should be stressed here that the fabrication of our monolithic metamaterial nanocavities does not require any additional processing steps. The complex flip-chip process [38, 39] that is normally required to realize a conductive ground plane can be avoided completely. Similar to the procedure used in the FDTD simulations we change the metamaterial resonance by geometric scaling and sweep it across the fixed IST. The normal incidence reflectance curves for the conventional sample are presented in Fig. 4(b). For this sample, we calculate a Rabi frequency of 2.1 THz following the same procedure as in the FDTD simulations. The monolithic metamaterial nanocavity results are shown in Fig. 4(c) and we demonstrate an increased Rabi frequency of 2.5 THz. As predicted by our FDTD simulations we were able to use the full three-dimensional confinement to maintain the same light-matter interaction strength while reducing the thickness of the QW stack. The reduced contrast in our reflectance measurements compared to the simulations is attributed to the use of a microscope objective with 0.58 NA leading to a large spread of incoming angles while the calculations are performed for normal incidence plane wave excitation. At the same time, the microscope objective allows us to sample an area of only $100 \times 100 \mu m^2$ and thereby minimize the influence of processing imperfections and heterostructure variations which would otherwise lead to a broadening of the measured resonances. The overall red-shift of our intersubband resonances for both samples compared to the calculations is attributed to a small thickness gradient across the wafer (center vs. edge).

One major difference between the two samples that was also observed in the FDTD simulations is the interchange of regions of high and low reflectance. The conventional sample shows a peak in reflectance on the metamaterial resonance and low background reflectance that is given by the refractive index contrast between air and the semiconductor. The monolithic metamaterial nanocavity shows a reflectance coefficient close to unity at off-resonance frequencies which is caused by the quasi-metallic ground plane. On resonance, the reflectance is reduced, showing the polariton eigenfrequencies as minima. This behavior is maintained below the bulk plasma frequency of the ground plane. When approaching 30 THz the real part of the permittivity of the n + layer approaches zero with a concomitant weakening of the “metallic character” of this layer. As a result this 800 nm layer becomes partially transmissive. Hence, the efficiency of the n + layer to act as a metallic layer and provide mode confinement is reduced.

4. Conclusion and outlook

In conclusion, we have presented a monolithic method to confine light in all three spatial dimensions using metamaterial nanocavities in conjunction with a semiconductor-based ground plane. The combination of a two-dimensional metamaterial with a buried n + layer leads to a reduced QW-stack thickness while maintaining the same light-matter interaction strength compared to a conventional metamaterial sample without ground plane. Our structures are compatible with planar processing technologies and can be realized in any frequency region as long as the IST remains below the bulk plasma frequency of the semiconductor ground plane. The reduction in the QW region thickness also means that the cavity volume is further reduced compared to a conventional metamaterial resonator. We have calculated a mode volume of $1.34 \times 10^{-3} \lambda_{\text{eff}}^3$ for the monolithic nanocavity sample which corresponds to a reduction by a factor of 1.9 as compared to a conventional metamaterial resonator making our monolithic nanocavities an excellent tool to study light-matter interaction in extremely small volumes. Our confinement concept can be extended towards higher frequencies by carefully engineering the n + layer. The strong light-matter interaction itself relies on ISTs which can be realized in various semiconductor
heterostructures. The plasma frequency of the ground plane can be increased by using semiconductors that allow for higher doping levels or have higher carrier mobility. The highly scalable device concept presented in this work is well suited for the realization of actively tunable metamaterials. The IST properties such as transition energy or dipole matrix element can be easily controlled by an external bias using the quantum-confined Stark effect [40, 41]. Due to the strong interaction between the IST and the monolithic metamaterial nanocavity also the response of the coupled system will be tuned. This leads to an active control of the metamaterial resonance by an external bias. The reduction in QW-stack thickness due to the additional mode confinement provided by the ground plane reduces the amount of active medium and thereby simplifies device tuning.

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