Ultra-Strong Light-Matter Interaction with Mid-Infrared Metamaterials

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Abstract: We present ultra-strong light-matter interaction of a metamaterial mode and an intersubband transition for normal incidence radiation in the mid-infrared spectral region. The anti-crossed lines show a splitting of 15\% of the central frequency.

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1. Introduction

Strong and ultra-strong light matter-interaction are highly interesting fields of research. If an optical cavity is coupled efficiently to a two-level system (the coupling is stronger than all the loss mechanisms) then energy is transferred from the cavity light field to the two-level system and back. If the exchange rate (called Rabi frequency) becomes similar to the fundamental cavity oscillations, then the system moves to the ultra-strong regime. One of the immediate outcomes is the non-classical ground state of the coupled system that becomes a “squeezed vacuum” containing a finite number of virtual photons [1]. These photons can be released in theory if the light-matter interaction is turned off non-adiabatically (much faster than the fundamental system resonance).

Here, we present an experimental study of ultra-strong coupling using a two-dimensional metamaterial (often called metasurface) as a sub-wavelength optical cavity in the mid-infrared (MIR) spectral region. For the two-level system we use intersubband transitions (ISTs) in semiconductor quantum-wells. The metamaterial resonators play a crucial role for optical coupling. Their near-field is polarized predominantly perpendicular to the semiconductor substrate (parallel to growth direction of the quantum-wells). This is the only polarization that can interact with intersubband transitions in conventional quantum-wells [2].

![Fig. 1](image)

**Fig. 1:** (left) Unit-cell of our metamaterial resonator. The optical dipoles (ISTs) are placed directly in the near-field of a “dogbone” metamaterial resonator. (right) Transmission spectra for different metamaterial resonances fabricated on top of the quantum-well sample (with an intersubband transition at 8.7 \( \mu \text{m} \)). The two systems couple and the splitting into the two polariton branches emerges as soon as the IST and the metamaterial resonance become similar in energy.

2. Experimental results and discussion

The metamaterial layer used in this work is based on an array of “dogbone”-resonators [3]. The schematic of one unit-cell is presented in Fig. 1(left). The “dogbone” shows a larger decay length of its cavity mode into the semiconductor compared to a conventional split-ring resonator. This allows us to couple more quantum-wells to the cavity mode and enhance the splitting. We observe ultra-strong light-matter interaction in the MIR range between 8 and 12 \( \mu \text{m} \). The optical transmission presented in Fig. 1(right) is measured for a quantum-well with its fundamental transition at 8.7 \( \mu \text{m} \) (140 meV, 34 THz). By varying the resonator size, the metamaterial resonance is swept across the IST which allows us to map out the two polariton branches. On resonance we observe a polariton splitting of 4.2
THz, corresponding to 12% of the center frequency. The other two quantum-well samples fabricated show ISTs at 10 and 12 μm respectively. We are able to increase the observed splitting to 15% of the center frequency. All experiments are performed at room-temperature as the ultra-strong coupling remains stable at elevated temperatures.

To verify the ultrafast energy exchange between the metamaterial and the IST we also measured the Rabi oscillations directly using time-domain spectroscopy. The experimental setup is based on a parametrically amplified, ultrafast fiber laser system seeded by a single ultrashort Er:fiber laser [4]; a schematic is presented in Fig. 2(left). The broadband MIR pulse is generated by difference frequency generation between two ultra-fast near-infrared pulses inside a GaSe crystal. The amplitude and phase of the pulse are mapped by electro-optic sampling inside a second, phase-matched GaSe crystal. We can clearly observe an oscillation of the cavity field with a period 33 fs, corresponding to the carrier frequency of 33 THz. On top of that we see an additional beating with a period of 480 fs which corresponds to a Rabi frequency of 2.1 THz (Fig. 2(right)). The MIR pulse has a pulse-energy of 0.1 pJ ensuring that we only probe the system in the weak excitation regime. We will further discuss a new strong coupling formalism that takes into account many body effects and strong exciting light fields.

![Fig. 2: (left) Schematic of the time-domain spectroscopy system used [4]. The ultrafast fiber laser system generates three near-infrared pulses that are phase-locked to a master oscillator. Two of those pulses are used to generate the broadband MIR pulse by difference frequency generation. The third pulse is used to measure amplitude and phase of the electric field by electro-optic sampling. (right) MIR pulse modified by the interaction with the strongly coupled light-matter system. The beating period of 480 fs corresponds to a Rabi frequency of 2.1 THz.](image)

3. Conclusion

In summary, we have presented ultra-strong light-matter interaction in the mid-infrared spectral region. We used a subwavelength metamaterial resonator as our cavity, and the matter dipoles were realized using inter-subband transitions in quantum wells. The splitting between the upper and lower polariton branches reaches 15% of the fundamental system resonance. Furthermore, we have also measured the Rabi oscillations directly in the time domain. A clear beating signal with a period of 480 fs has been observed.

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4. References

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