Deep-Strong Coupling between Cavity Photons and Terahertz TO Phonons in PbTe

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Abstract: We have investigated the resonant coupling of photons with TO phonons in lead telluride in small-mode-volume terahertz cavities, observing a giant vacuum Rabi splitting on the order of the bare cavity–phonon frequency.

1. Introduction

There is currently much interest in searching for the novel states, phases, and phenomena that are predicted to exist in new regimes of strong light–matter interaction [1–3]. The ultrastrong coupling (USC) regime arises when the light–matter coupling strength, g, becomes a significant fraction of the bare resonance frequency of light and matter, ω_0. Furthermore, when g exceeds ω_0, the light–matter hybrid enters the deep-strong coupling (DSC) regime. The DSC regime has been achieved in multiple experimental platforms, e.g., superconducting circuits [4], Landau polaritons in a two-dimensional electron gas [5], and plasmonic crystals [6]. However, much of the new exotic physics expected to occur in the DSC regime remains unexplored.

Here, we present a novel experimental system in which to investigate DSC physics: terahertz (THz) cavity phonon polaritons. Specifically, we study strongly anharmonic transverse optical (TO) phonons in lead telluride (PbTe) films [7] that are resonantly coupled to THz photons in small-mode-volume metasurface cavities. At zero detuning, we observed a vacuum Rabi splitting whose value ($\Omega = 2g$) is close to twice the cavity–phonon bare frequency ($2\omega_0$), reaching the DSC regime. We systematically studied the coupling strength as a function of sample thickness, temperature, and cavity length. The obtained experimental results were in agreement with results of electromagnetic simulations we performed.

2. Methods

We studied single-crystalline PbTe films on (111) BaF_2 substrates grown by molecular beam epitaxy. We used THz time-domain spectroscopy to measure the TO-phonon resonance and determined the optical constants of the films. We then used the determined optical constants to define the material parameters for simulating the coupling of the phonon resonance with the metasurface cavity using the CST Studio Suite. The cavity design is shown in the Fig. 1, with the THz vacuum photons confined in the gaps of the I-shaped metasurface that couples to the TO phonon mode of PbTe.

We used maskless photolithography to fabricate a metasurface structure. The uncoupled resonance was characterized for a metasurface/Si sample. Figure 1(a) shows our metasurface cavity design. It provides electric field confinement in 4 µm gaps, as shown in Fig. 1(b). Our design allows easy fabrication of such cavities in terms of resolution due to large enough features suitable for maskless photolithography. Figure 1(c) shows an optical image of one of the samples we fabricated.

To characterize the cavity and light–matter coupling, we used a home-built THz time-domain spectroscopy setup in transmission geometry. A broadband THz pulse (0.25–2.5 THz) was generated by a photoconductive

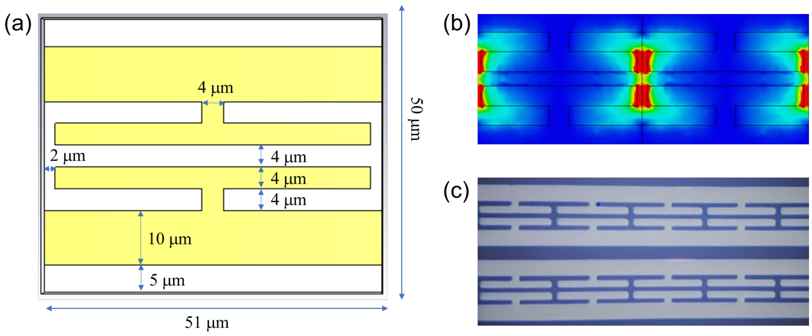

Fig. 1. (a) Schematic diagram for our metasurface cavity design. (b) Electric field distribution as obtained by numerical simulations. (c) An optical image of the fabricated metasurface.

antenna and detected via electro-optic sampling in a ZnTe crystal using 800 nm pulses with a duration of 150 fs and a repetition rate of 80 MHz from a Ti:sapphire oscillator.

3. Results and Discussion

Figure 2(a) shows a transmittance spectrum for a metasurface deposited onto a 300-nm-thick PbTe sample (red solid line). The data agrees well with simulation results shown by the blue dashed line. The uncoupled cavity resonance and the TO phonon mode of PbTe are at 0.91 THz. The experimental and numerical data show two pronounced peaks, which we attribute to the lower and upper polaritons. The Rabi splitting, $\Omega = 2g$, between these polaritons is 1.6 THz. The normalized coupling strength is, therefore, $\eta = \Omega/2\omega_0 \approx 0.9$.

Further calculations as a function of PbTe thickness shown in Fig. 2(b) place the system into the DSC regime. The dots represent results of numerical simulations, while the solid line represent a fit with a square root function. These results indicate that this platform is promising for realizing and understanding cavity-vacuum-induced ferroelectric phase transitions, as well as for exploring applications of the USC/DSC regimes in quantum technology.

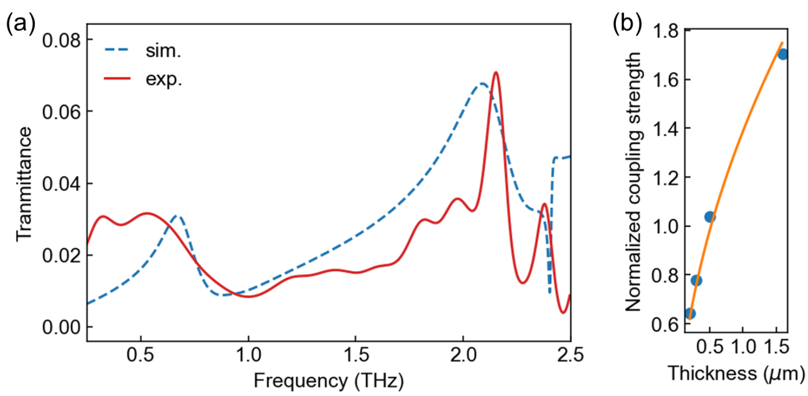

Fig. 2. (a) Simulated and measured transmittance spectra for a 300-nm-thick PbTe sample with a designed meatsurface. (b) Normalized coupling strength as a function of PbTe thickness obtained from numerical simulations.

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