Nonlocal Coherent Optical Nonlinearities of a Macroscopic Quantum System

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The optical responses of solids are typically understood to be local in space. Whether locality holds for the optical response of a macroscopic quantum system has, however, been largely unexplored. Here, we use multidimensional coherent spectroscopy at the optical diffraction limit to demonstrate nonlocal optical nonlinearities in a semiconductor microcavity. These nonlocal optical responses are both coherent and quantum in nature, deriving from the macroscopic length scale of confined exciton-polariton wave functions.

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In standard treatments of light-matter interactions, the material response is assumed to be local in space. Despite its ubiquity, the limitations of this assumption are of both fundamental and practical importance. For example, it is well known that optical responses can exhibit nonlocality when length scales reach the near-field regime [1–3]. Nonlocal optical responses can also occur when mobile excitations travel to other points in space for example via photorefraction [4], thermal [5] and charge diffusion [6], or phonon-polariton propagation [7]. To manifest any of the above effects that rely on energy or charge transport, however, the optical medium need not be quantum (i.e., no macroscopic quantum coherence is necessary).

Theoretical predictions for specially prepared atom-waveguide systems [8] have pointed to quantum effects as a potential new source of nonlocal optical responses. Yet intuitively, any macroscopic quantum state should naturally exhibit nonlocal optical responses by virtue of a delocalized wave function. In real materials, however, short coherence lengths typically preclude observation of such nonlocality. Exciton polaritons, hybrid excitations between electron-hole pairs and light [9], are an exception to this conventional wisdom. With coherence lengths reaching macroscopic length scales, spectacular demonstrations of quantum effects have been reported, ranging from Josephson oscillations [10,11] to Bose-Einstein condensation [12,13].

As composite light-matter particles, polaritons are also amenable to photonic engineering techniques. For example, exciton polaritons in a semiconductor microcavity can be laterally confined by sharp discontinuities in the cavity refractive index. An elegant way this has been accomplished is by use of a subwavelength grating (SWG) as a cavity mirror, in which the grating dimensions impose an effective confinement potential [14,15]. The resultant micrometer length-scale exciton-polariton wave functions are a perfect host for *nonlocal* nonlinear optics that rely on macroscopic quantum coherence rather than transport effects, which we demonstrate in this Letter.

The system studied is illustrated in Fig. 1(a), composed of GaAs quantum wells (QWs) embedded in a cavity formed by $Al_{0.15}GaAs/AlAs$ distributed Bragg reflector (DBR) layers and a square SWG that acts as a high reflector for light polarized along the grating bars [referred to here as transverse electric (TE) polarization]. The TE-polarized photoluminescence reveals three-dimensional quantum confinement of the exciton polaritons, as shown by the spectrally resolved real-space image in Fig. 1(b). The lower polariton branch is split into discrete eigenstates due to the lateral confinement, whose energies and spatial distributions are reproduced by a harmonic confinement potential [15]. The macroscopic polariton states observed (with spatial extent exceeding the corresponding optical diffraction limit) can now be selectively excited by positioning optical excitation at the antinodes of their wave function. For simplicity, here we restrict our attention to the two lowest-energy states indicated by the dashed lines in Fig. 1(b).

While the linear optical properties of these zero-dimensional polaritons are now well understood, their nonlinear optical properties remain unexplored. Here, we apply the technique of multidimensional coherent spectroscopy (MDCS), capable of characterizing the full multidimensional nonlinear optical response of a given system [16,17]. MDCS has been used to study many aspects of polaritonic physics, ranging from polariton coupling [18,19] to their higher-order excitations [20,21], but its application to quantum-confined polaritonic systems has not yet been reported.

A schematic of our MDCS experiment is shown in Fig. 1(c) in which three TE-polarized excitation pulses

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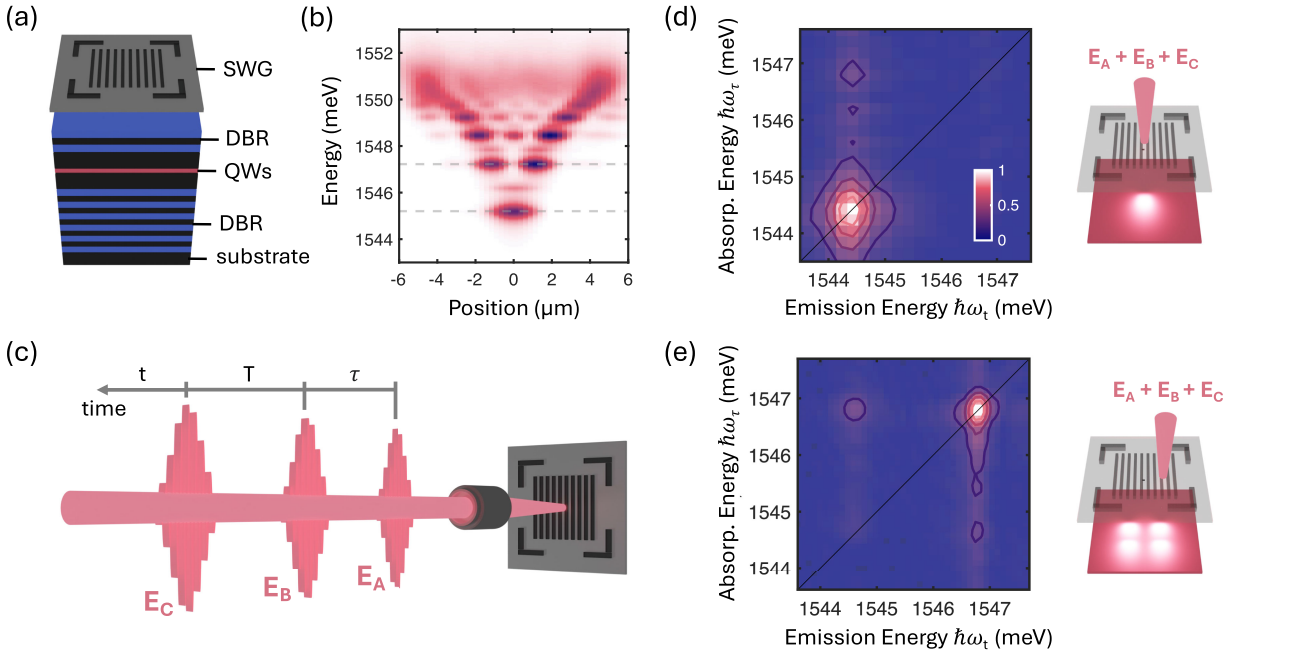

FIG. 1. (a) Schematic of the hybrid cavity composed of quantum wells (QWs) between distributed Bragg reflectors (DBRs) and a subwavelength grating (SWG) mirror. (b) Spectrally resolved real-space image of photoluminescence from the device in (a), showing the spatial profiles of the confined lower-branch polariton modes. Dashed lines marked the first and second excited states probed here. (c) Schematic of the MDCS measurement, in which three excitation pulses $\{E_A, E_B, E_C\}$ are focused by a microscope objective ($NA = 0.5$) on the sample. The nonlinear signal is measured as a function of time delays $\{\tau, t\}$ with $T = 200$ fs for all measurements. (d),(e) Single-quantum spectra with the excitation beams positioned at the (d) center of the SWG (antinode of the first excited state) and (e) 1.5 μm to the right and 0.5 μm below (antinode of the second excited state). Cartoons depict the excitation beams targeting specific exciton-polariton wave functions in the quantum wells (red squares).

$\{E_A, E_B, E_C\}$ (with sufficient bandwidth to excite all of the states identified by photoluminescence) impinge on the sample at normal incidence, cooperatively generating a coherent electric field as a function of the time delays $\{\tau, T, t\}$ as indicated. We note that the collinear excitation geometry used here [18,22,23] (in contrast to the more common noncollinear geometry [16]) is crucial to reaching a near diffraction-limited excitation spot size (≈1.1 μm). By measuring the nonlinear electric field along two time delays and performing a two-dimensional (2D) Fourier transform, the dynamics along each time delay are correlated in a 2D spectrum. Various types of 2D spectra are possible [16] that reveal unique aspects of a system's microscopic physics, but here we focus on the spectrum obtained by Fourier transforming along the delays $\{\tau, t\}$. These single-quantum spectra correlate the photon energies of initial optical absorption and subsequent optical emission in a nonlinear wave-mixing process, which are ideal for measuring both coherent and incoherent coupling between optical transitions. Note that, due to minimal inhomogeneous broadening, the two types of single-quantum spectra (rephasing and nonrephasing) provide identical information (see Supplemental Material [24] for comparison). We therefore consider nonrephasing spectra in the following, due to a slight advantage in peak clarity.

To demonstrate the spatial selectivity of confined exciton-polariton wave functions, single-quantum spectra acquired with excitation at two distinct sample positions are plotted in Figs. 1(d) and 1(e). In the spectrum shown in Fig. 1(d), all excitation beams are positioned at the antinode of the first excited-state mode (center of the SWG). A single peak is observed that arises from optical absorption (vertical axis $\hbar\omega_\tau$) and optical emission (horizontal axis $\hbar\omega_t$) at the first excited-state polariton energy. In the spectrum shown in Fig. 1(e), the excitation beams are translated (1.5 μm laterally and 0.5 μm vertically away from the center position) to an antinode of the second excited-state mode. A single dominant peak is again observed, this time arising from optical absorption and optical emission at the second excited-state polariton energy.

Having demonstrated that each exciton-polariton wave function offers a locally distinct nonlinear optical response, we now investigate the possibility of a nonlocal nonlinear optical response *between* different wave functions. Referring to the first two excitation pulses $(E_A + E_B)$ as the pump and the third excitation pulse (E_C) as the probe, spectra acquired with the pump and probe pulses separated in space are plotted in Fig. 2. For the spectrum in Fig. 2(a), the pump pulses are spatially centered at the antinode of the first excited-state while the probe pulse is centered at that of

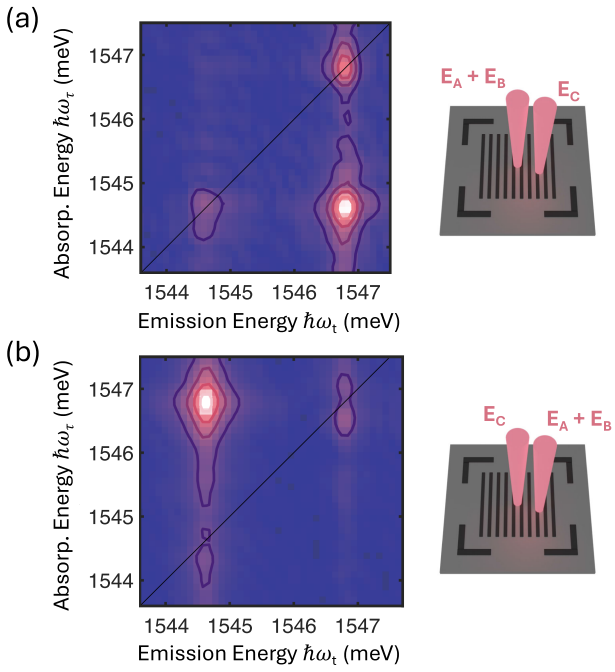

FIG. 2. (a),(b) Single-quantum spectra acquired with the "pump" ($E_A + E_B$) and "probe" (E_C) pulses exciting different spatial locations on the sample. (a) The (probe)pump pulses interact with the (second)first excited state. (b) Spatial locations of the pump and probe pulses are exchanged.

the second excited state. With this excitation scheme a new peak now emerges whose position corresponds to absorption at the first excited-state polariton energy and emission at the second excited-state polariton energy, direct evidence of nonlocal optical nonlinearities between two spatial quantum wave functions. By exchanging pump and probe positions, the inverse process of absorption at the second excited-state energy and emission at the first excited-state energy is observed in the spectrum in Fig. 2(b). These observations demonstrate that the coupling we observe via MDCS is not simply energy relaxation between the two states, but is indeed true coherent coupling in which optical absorption by one resonance modifies the optical response of the other at a different spatial location.

To confirm that the observed local and nonlocal nonlinearities indeed derive from the spatial-dependence of distinct exciton-polariton wave functions, we perform simulations of the single-quantum MDCS spectra (see Supplemental Material [24]) based on a standard perturbative solution of the system density matrix dynamics [16]. While full electrodynamic simulations of each two-dimensional exciton-polariton state were prohibitively difficult, the spatial-dependences of their transition dipole moments can be well estimated from their respective photoluminescence intensity profiles in Fig. 1(b). For each exciton-polariton state, the square root of their intensity profile is used as a proxy of the local transition dipole moment and is assumed to be symmetric along and across the grating bars.

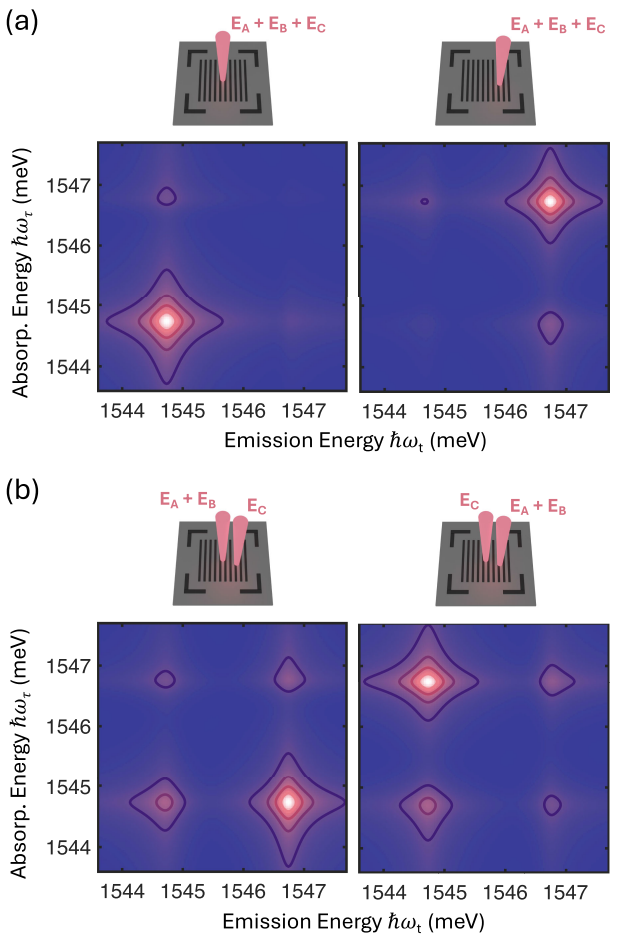

FIG. 3. Perturbative density matrix simulations of (a) local and (b) nonlocal exciton-polariton nonlinearities. We use local dipole moments estimated from the real-space photoluminescence intensity in Fig. 1(b) and assumed dephasing rates of 0.3 and 0.25 ps^{-1} for the lower- and higher-energy states, respectively.

We further account for the finite spatial extent of optical excitation by averaging each local dipole moment over a Gaussian beam profile of 1.1 μm full-width at half max (see Supplemental Material [24]), and assume 0.3 and 0.25 ps^{-1} dephasing rates for the lower- and higher-energy states, respectively. The resultant simulations are plotted in Fig. 3, which agree well with the corresponding experimental spectra shown in Figs. 1 and 2. We note that to achieve this good agreement, one must account for the heterodyne detection process. In short, the local oscillator pulse is aligned to the probe pulse and interferes optimally with signals that match the spatial mode of E_C, thereby emphasizing signals emitting from the first excited state in the excitation scheme of Figs. 1(d) and 2(b) while emphasizing signals emitting from the second excited state in that of Figs. 1(e) and 2(a). We emphasize, however, that the spectral features observed in Fig. 2 are still dominated by the nonlocal nonlinear optical responses (see Supplemental Material [24] for further discussion and simulations that disregard heterodyne detection efficiency).

Accounting for the finite excitation beam profiles also captures the residual "on-diagonal" ($\hbar\omega_\tau = \hbar\omega_t$) peaks, which could be minimized by improving the spatial excitation scheme, increasing the length scale of the exciton-polariton wave functions [15], and/or decreasing the wavelength of the excitation light (smaller diffraction limit). Finally, population relaxation and higher-lying states are neglected in the simulations, but do not contribute to the salient physics considered here.

In conclusion, we demonstrate nonlocal optical nonlinearities between two distinct exciton-polariton wave functions. This demonstration of a nonlocal coherent optical response in a macroscopic quantum system raises possibilities of nonlocal optical nonlinearities in other quantum material platforms, for example in superconductors with terahertz optical responses [25]. Here, we considered third-order nonlinearities between two polaritonic states, but the possibilities expand dramatically by considering higher-order nonlinearities and higher-energy wave functions. With further refinement of the technique presented here, we envisage the generation of designer spatial emission patterns using these nonlocal nonlinearities and a new paradigm of coherent *spatial* control.

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Data availability—The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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