Two-photon injection of polaritons in semiconductor microstructures

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We experimentally demonstrate that two-photon pumping of "dark" excitons in quantum wells embedded in semiconductor microcavities can result in exciton-polariton injection and photon lasing. In the case of a semiconductor micropillar pumped at half of the exciton frequency, we observe a clear threshold behavior, characteristic of the vertical cavity surface emitting laser transition. These results are interpreted in terms of stimulated emission of terahertz photons, which allows for conversion of "dark" excitons into exciton-polaritons.

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Terahertz radiation (electromagnetic radiation with frequencies between the infrared and microwave range of the spectrum) is scarcely explored to date. This energy range has many useful properties, such as nondestructive penetration in common materials and biological tissues. There is currently a strong demand for development of new compact and efficient terahertz sources for numerous applications in sensing, communications, imaging in biology and medicine, material science, security, and ultrafast computing to name but a few [1]. Emitters of terahertz radiation based on nonlinear-optical frequency downconversion have been around for years but they are bulky, impractical, and expensive. More compact quantum cascade lasers [2] remain short-living and extremely expensive. Recently, the physical effect of Bose–Einstein condensation has been proposed as a tool for realization of a new generation of lasers: bosonic lasers [3]. In these devices the emission of terahertz photons can be amplified by orders of magnitude due to the stimulation of radiation by a coherent many-body state: the bosonic condensate. A bosonic terahertz laser will have to be combined with a conventional light-emitting diode to provide an efficient pumping scheme based on the two-photon absorption (TPA). TPA is defined as the simultaneous absorption of pairs of photons of the same or different energy in order to excite an electronic transition. This phenomenon was first predicted by Göppert-Mayer in 1931 [4], and demonstrated 30 years later by Kaiser and Garret [5] by illuminating a crystal of CaF_2 containing Eu^{2+} ions with a ruby laser beam. TPA is nowadays a widely diffused technique in the fields of chemistry, biology, and photonics [6–9]. It is used in various types of quantum confined light emitters, such as single molecules [6] or colloidal nanocrystals [10,11]. Recently, TPA attracted the attention of the scientific community in the field of exciton-polaritons [12,13]. Indeed, TPA has been theoretically proposed as a new injection mechanism for these mixed light-matter particles in semiconductor microcavities [14]: pumping at half of the frequency of the 2p exciton state one can create a reservoir of optically forbidden ("dark") excitons, which can be subsequently converted to exciton-polaritons with emission of terahertz photons [15,16].

It is worth mentioning that the injection of exciton-polaritons by TPA would be allowed only if the two-photon beam is suitable to inject excitons in the active materials composing the system. In planar microcavities the active part usually consists of one or more quantum wells. Excitons are elementary crystal excitations, which may or may not be excited by light depending on their spin and orbital quantum numbers. In zinc-blend semiconductors (e.g., GaAs) the s-exciton states (including the lowest energy 1s-exciton state) are optically allowed, while the excited (p,d,f) states are forbidden (they are referred to as "dark" excitons) due to violation of the total angular momentum conservation rule. Nevertheless, these states can be excited by multiphoton pumping, where each photon carries an angular momentum equal to 1. In particular, it is well known that p-excitons may be created by TPA [4,17], each photon having one half of the p-exciton energy. Thus, TPA allows for selective excitation of p-exciton states [18,19].

This work reports the first experimental demonstration of two-photon polariton injections in semiconductor microcavities by using femtosecond laser pulses centered between two polariton branches. We have tested this technique in two different systems: semiconductor planar microcavities (with both excitons and photons confined in the growth direction) and pillar microcavities (having three-dimensional photons confinement). In the

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case of micropillars, the photon lasing regime is clearly observed and, as described below, it paves the way toward the realization of vertical cavity surface emitting terahertz lasers.

The investigated structures were cooled at 4 K using a cold-finger cryostat and excited by means of a femtosecond pulsed laser beam (pulse duration ≈150 fs, full width at half-maximum $\Delta E_{pump} \approx 13$ meV) with an energy E_{pump} chosen following the requirements of the specific experiments, as described in the next paragraphs. As a general rule, the two-photon injection technique consists in the excitation of the polaritons at energies of $E_{pump} = E_P/2$, with E_P the energy of the exciton state to be excited. The laser beam, provided by an optical parametric oscillator pumped by a femtosecond pulsed Ti:sapphire laser, was focused on the sample by an aspherical lens (beam diameter of about 30 μm at waist). The resulting photoluminescence (PL) was collected through the same optics and the reflected laser beam filtered out by a dichroic mirror. The signal was dispersed in a 500 mm long spectrometer in order to perform real-space and far-field analysis.

We will first analyze the PL properties of a two-photon excited planar microcavity consisting of three $In_{0.04}Ga_{0.96}As$ quantum wells (QWs) placed at the center of a 2λ-cavity composed by GaAs/AlGaAs Bragg mirrors (21, 24 pairs, respectively, front and back), with a Rabi splitting of 5.8 meV and polariton eigenstates E_{LP} ($k_\parallel$) and E_{UP} ($k_\parallel$) in the energy interval [1.47 eV; 1.495 eV] ($k_\parallel$ is the wave vector projection in the microcavity plane).

It is worth mentioning that the spectral width of the pulsed pump, ΔE_{pump}, is larger than the vacuum Rabi splitting of the investigated systems and, when the cavity-exciton detuning $\delta_{C-X} = E_C - E_X$ approaches zero (with E_C the resonant energy of the cavity and E_X the energy of the exciton), both the lower and upper polariton branches are excited. For this reason, in each case the pump energy will be compared to the average value of the lower and upper polariton energy at $k_\parallel = 0$, i.e., $E_{ave} = [E_{LP}(k_\parallel = 0) + E_{UP}(k_\parallel = 0)]/2$.

When the pump is tuned at energies $2E_{pump} \gg E_{ave}$, polaritons are injected out of resonance, and they are allowed to spontaneously relax exploiting the electronic reservoir. Indeed, in the detected dispersion curve, continuous upper and lower polariton branches are visible [Fig. 1(a)], thus confirming the strong exciton–photon coupling. In order to prove that polaritons have been injected by a TPA process, the PL intensity (I_{PL}) dependence as a function of excitation power (P_{pump}) for three different values of $k_\parallel$ has been fitted by the function $I_{PL} = bP^{a}_{pump}$, displayed in Fig. 1(b). All analyzed curves showed a quadratic dependence (i.e., a ≈2) of the emission intensity on the pumping intensity for both up and low polariton branches, as expected for an efficient TPA [4].

For $2E_{pump} = E_{ave}$, we have instead observed that polaritons are injected conserving the momentum of the pump in the transverse plane, similarly to the standard resonant excitation [12]. Indeed, the PL intensity reported in Fig. 2(a), obtained for $\delta_{C-X} \approx 0$, shows strong emission at the injection momentum $k_\parallel = 0$. Like in the previous case, polaritons can spontaneously relax to all states of the lower branch, which explains the emission of the whole lower polariton branch. Also in this case, quadratic dependence of PL intensity on excitation power for the detected signal has been observed at each investigated detection $k_\parallel \neq 0$ [see Fig. 2(b)], although with much lower intensity, due to the lack of momentum conservation in polaritons relaxation. Actually, when the in-plane momentum of the excitation laser beam is modified, the maximum detected PL intensity moves along the dispersion curves accordingly to the excitation wavevector $k_\parallel$, as shown by the lower branch dispersion curves displayed in Fig. 3, thus demonstrating that the

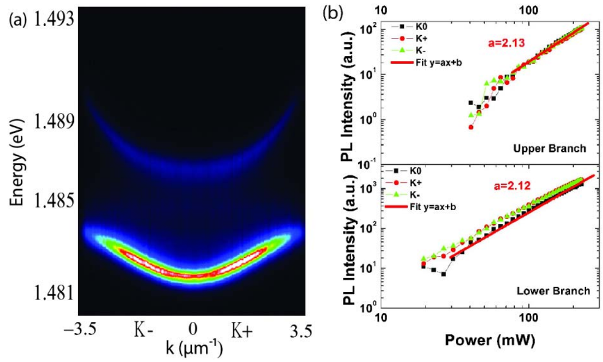

Fig. 1. Planar microcavity pumped out of resonance: $2E_{pump} \gg E_{ave}$. (a) Dispersion curve of the planar microcavity with $E_{ave} = 1.485$ eV for an excitation energy of $E_{pump} = 0.780$ eV and $P_{pump} = 100$ mW. (b) Power dependence of the upper and lower branch for different in-plane momenta. The dashed red line shows the fitting of the power dependence of the emitted intensity with the function $I_{PL} = bP^{a}_{pump}$. The fitting was done for three different momenta: K− = −2 μm^{-1}, K0 = 0 μm^{-1}, and K+ = 2 μm^{-1}.

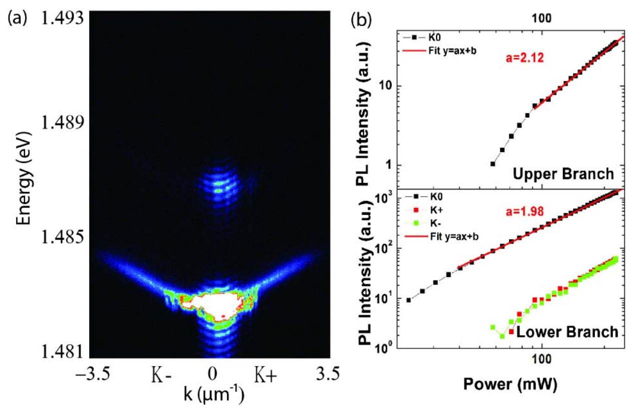

Fig. 2. Two-photon resonant excitation of a planar microcavity: $2E_{pump} = E_{ave}$. (a) Dispersion curve of the planar microcavity with $E_{ave} = 1.485$ eV for an excitation energy of $E_{pump}(k_\parallel) = 0.743$ eV and $P_{pump} = 100$ mW. The lines visible around $k = 0$ are due to a parasitic interference due to a spurious reflection of the emitted beam on the optical elements in the excitation setup. (b) Power dependence of the upper and lower branch for different in-plane momenta. The dashed red line shows the fitting of the power dependence of the intensity with the function $I_{PL} = bP^{a}_{pump}$. The fitting was done for three different momenta: negative K− = −2 μm^{-1}, zero K0 = 0 μm^{-1}, and positive K+ = 2 μm^{-1}.

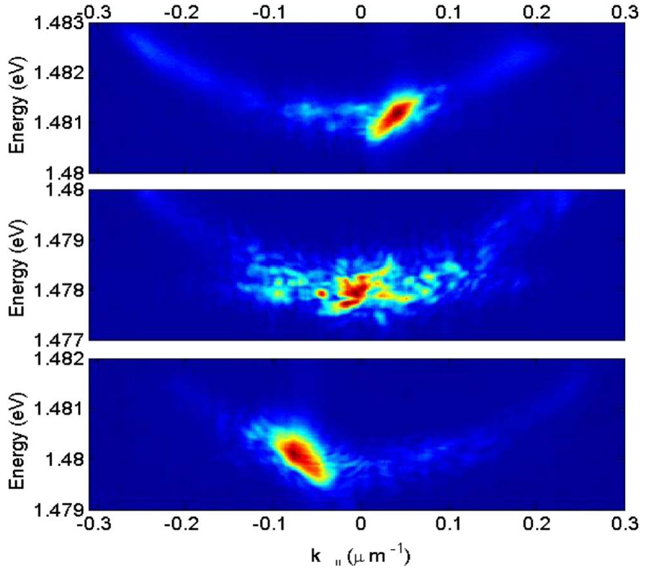

Fig. 3. Dispersion curves for three different injection momenta with two-photon resonant excitation ($2E_{pump} = E_{ave}$) of a planar microcavity.

pumping condition $2E_{pump} = E_{ave}$ is equivalent to a resonant injection of polaritons.

We have thus shown that two-photon excitation is an efficient technique to inject polaritons in a planar microcavity. In order to extend the proposed technique to other systems, we have replicated the experiment on circular pillar microcavities with diameters and quality factors ranging from 2 to 15 μm and from 4000 to 6000, respectively. These structures are composed by 26 and 30 pairs of $Ga_{0.9}Al_{0.1}As/Ga_{0:05}Al_{0.95}As$ Bragg mirrors for top and bottom reflectors, respectively, having a single $In_{0.05}Ga_{0.95}As$ QW between them. This design is chosen in order to show that the TPA is efficient also in a fully confined optical system. The λ-cavity is characterized by a Rabi splitting of 3.5 meV. The excitation beam was focused by means of the same optical path as for the experiments on the planar microcavity complemented by a microscope objective with a NA of 0.7, resulting in a spot size of ~3 μm on the sample. In this case the high optical confinement introduced by the pillar microcavity and the small focalization spot, needed to match the pillar size, are expected to increase the polariton–polariton interactions with respect to the case of planar microcavities.

It is well known that once the number of polaritons exceeds a certain threshold the strong coupling is broken and the system enters in the weak-coupling regime [20]. Since TPA is based on a femtosecond pulsed excitation, the number of injected polaritons is not constant in time and, during the same pulse, weak and strong coupling can coexist in the pillar. To properly analyze the PL emission, a time-resolved detection system was used. In order to simultaneously record temporal and energy evolution of micropillars PL, a second 500 mm long spectrometer was coupled to a streak camera (Hamamatsu, Model C5680, with time resolution of 5 ps) and time-resolved emission spectra for several micropillars were registered as a function of the excitation power. In the experiments described below the pumping laser energy has been tuned in order to obtain the best possible excitation of the lowest polariton states of both upper and lower branches.

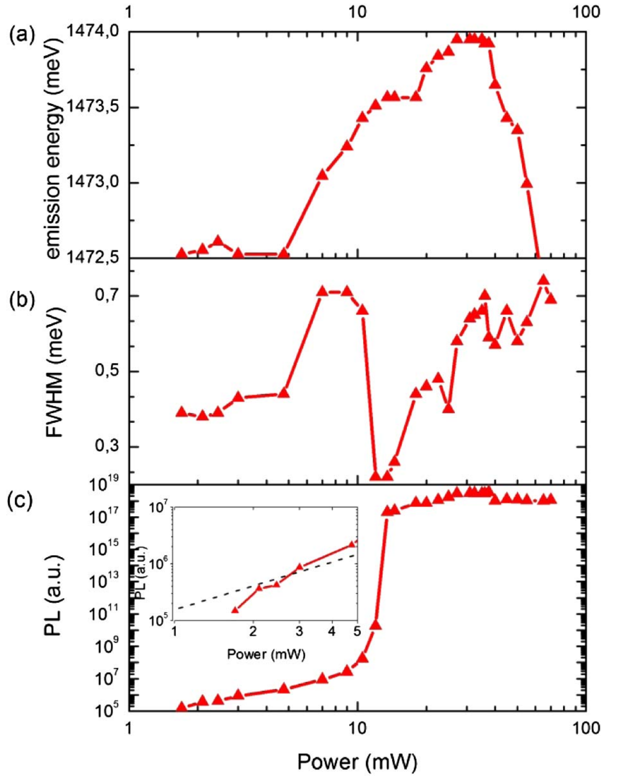

Fig. 4. Dependence of micro-pillars (MPs) photoluminescence on the power of the excitation laser beam for a pillar of a 4 μm diameter. (a) Emission energy as functions of the excitation power, (b) full width at half-maximum (FWHM), and (c) PL intensity as a function of excitation power. The inset of panel (c) shows a zoom for low power excitation. Black-dashed line represents the function $I_{PL} = bP^{a}$ with $a = 2$.

A representative PL intensity of the ground state of a pillar microcavity as a function of the excitation power is displayed in Fig. 4(c): it clearly shows a threshold, below which the PL intensity depends quadratically on the excitation power, showing the effectiveness of TPA also in pillar microcavities. Notably, an emission energy blueshift due to the increasing number of polaritons in the system has been observed [see Fig. 4(a)]. At threshold, the ground-state linewidth decreases [see Fig. 4(b)], the strong coupling breaks down, and the system enters into the photon laser regime. A saturation of the ground-state emission is observed for higher excitation power, accompanied by a redshift, which can be associated with the heating of the system due to high numbers of carriers injected by the excitation pulse [21].

In conclusion, we have shown that two-photon excitation is an efficient technique to inject polaritons in 1D and 3D confined structures. It allows injecting polaritons conserving the excitation momentum and obtaining the laser regime in pillar microcavities. According to recent theoretical predictions, the observed phenomena can be interpreted in terms of "dark" exciton injection followed by their conversion into "bright" exciton-polaritons assisted by terahertz photons emission, paving the way toward the realization of vertical cavity terahertz lasers.

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