Stimulated emission of terahertz radiation by exciton-polariton lasers

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We show that planar semiconductor microcavities in the strong coupling regime can be used as sources of stimulated terahertz radiation. Emitted terahertz photons would have a frequency equal to the splitting of the cavity polariton modes. The optical transition between upper and lower polariton branches is allowed due to mixing of the upper polariton state with one of the excited exciton states and is stimulated in the polariton laser regime.
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The realization of efficient terahertz radiation sources and detectors is one of the important objectives of the modern applied physics.[1,2] None of the existing terahertz emitters universally satisfies the application requirements. For example, the emitters based on nonlinear-optical frequency downconversion are bulky, expensive, and power consuming. Various semiconductor[3] and carbon-based[4,5] devices utilized intraband optical transitions that are compact but have a limited wavelength adjustment range and a low quantum efficiency. Among the factors that limit the efficiency of semiconductor terahertz sources is the short lifetime of the electronic states involved (typically, fractions of a nanosecond) compared to the time for spontaneous emission of a terahertz photon (typically milliseconds). The methods of reducing this mismatch include the use of the Purcell effect[6–8] in terahertz cavities or the cascade effect in quantum cascade lasers (QCL).[9] Nevertheless, until now, the QCL in the spectral region of about 1 THz remain costly and short-lived and still show the quantum efficiency of less than 1%. Recent studies of strong coupling intersubband microcavities[10] have shown a possibility of stimulated scattering of intersubband polaritons.[11] Here we explore the possibility of generating terahertz radiation in semiconductor microcavities in the regime of exciton-polariton lasing. The quantum efficiency of this source is governed by population of the final polariton state, which may be tuned over a large range by means of the optical pumping.

In the strong coupling regime in a microcavity,[12] the dispersion of the exciton polaritons is described by two bands both having minima at zero in-plane wave vector $\mathbf{k}$. At $\mathbf{k}=0$, the energy splitting between the two branches approximately equals $\hbar\Omega_R$, where $\hbar\Omega_R$ is the optical Rabi frequency, the measure of the light-matter coupling strength in a microcavity. Typically, $\hbar\Omega_R$ is of the order of several meV, which makes this system attractive for terahertz applications. Stimulated scattering of exciton polaritons into the lowest energy state leads to so-called polariton lasing, recently observed in GaAs, CdTe, and GaN based microcavities.[13,14] If the scattering from the upper to the lower polariton branch was accompanied by the emission of terahertz photons, polariton lasers would emit terahertz radiation, and this emission would be stimulated by the population of the lowest energy polariton state. However, at first glance, this process is forbidden since an optical dipole operator cannot directly couple the polariton states formed by the same exciton state.

This obstacle can be overcome if one of the polariton states of interest is mixed with an exciton state of a different parity, say, the $e1hh2$ exciton state formed by an electron at the lowest energy level in a quantum well (QW) and a heavy hole at the second energy level in the QW. This state is typically a few meV above the exciton ground state $e1hh1$. Nevertheless, by an appropriate choice of the QW width and exciton-photon detuning in the microcavity, the state can be brought into resonance with the lowest energy upper polariton state. Being resonant, the two states can be easily hybridized by any weak perturbation, such as, e.g., a built-in or applied electric field. The optical transition between such a hybridized state and the lowest $e1hh1$ exciton-polariton state is allowed.

We consider the model optical system shown at Fig. 1. It consists of a planar microcavity operating in the strong coupling regime, and placed inside a terahertz cavity, which additionally enhances the rate of the emission of the terahertz photons and can be constructed using various approaches

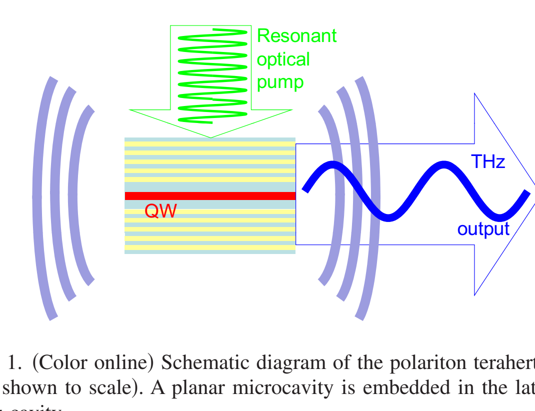

FIG. 1. (Color online) Schematic diagram of the polariton terahertz emitter (not shown to scale). A planar microcavity is embedded in the lateral terahertz cavity.

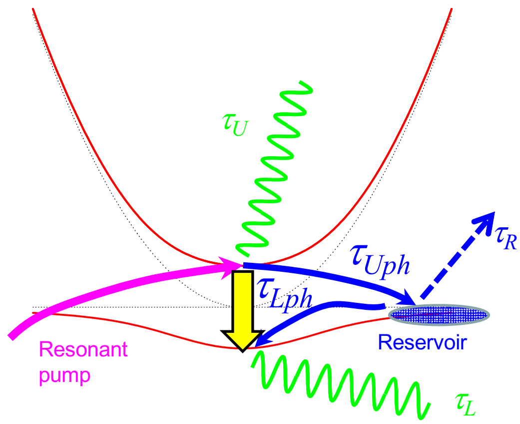

FIG. 2. (Color online) A schematic diagram illustrating possible transitions in the system. The vertical axis is the energy and the horizontal axis is the in-plane wave vector.

that are experimentally verified.[15,16] Together with the waveguiding effect of the microcavity structure, adding the lateral terahertz cavity would achieve an effective three-dimensional confinement of the terahertz mode, giving rise to enhancement of spontaneous emission rate through the Purcell effect.[6]

The eigenstates of the system can be obtained from the diagonalization of the Hamiltonian matrix, which is written on the basis of the bright exciton, dark exciton, and cavity photon (see the scheme in Fig. 2)

$$\hat{H} = \begin{vmatrix} E_{1X} & \delta/2 & \hbar\Omega_R/2 \\ \delta/2 & E_{2X} & 0 \\ \hbar\Omega_R/2 & 0 & E_C \end{vmatrix}, \qquad (1)$$

where E_{1X}, E_{2X}, and E_C are the energies of bright and dark excitons and the cavity photon mode, respectively, δ is a parameter describing the coupling between the bright and dark exciton states due to an electric field ε normal to the QW plane, $\delta \approx e\varepsilon \int_{-\infty}^{+\infty} u_{hh1}(z) u_{hh2}(z) dz$ with u_{hh1} and u_{hh1} being the confined wave functions of the ground and first excited states of the heavy hole in a QW. One can easily estimate that to have δ larger than the upper polariton linewidth, which is typically of the order of 0.1 meV, it is sufficient to apply an electric (or have a piezoelectric) field of 3 kV/cm in a 10-nm-wide quantum well. The diagonalization of the Hamiltonian (1) gives the eigenenergies and eigenstates of the system. In the case of $|E_{1X} - E_{2X} - \hbar\Omega_R/2| \ll \hbar\Omega_R$ and the zero detuning between bright exciton and photon modes $E_{1X} = E_c = E_1$, the eigenmodes are

$$|L\rangle \approx \frac{1}{\sqrt{2}}(|C\rangle - |1X\rangle), \qquad (2)$$

$$|U_+\rangle \approx \frac{1}{\sqrt{1+b_-^2}}\left[\frac{1}{\sqrt{2}}(|C\rangle + |1X\rangle) + b_-|2X\rangle\right], \qquad (3)$$

$$|U_-\rangle \approx \frac{1}{\sqrt{1+b_+^2}}\left[\frac{1}{\sqrt{2}}(|C\rangle + |1X\rangle) + b_+|2X\rangle\right], \qquad (4)$$

with $b_\pm = \delta^{-1}[E_{2X} - \hbar\Omega_R/2 - E_1 \pm \sqrt{(E_{2X} - \hbar\Omega_R/2 - E_1)^2 + \delta^2}]$. The notations $|L\rangle$, $|U_-\rangle$, and $|U_+\rangle$ correspond to the lower polariton branch and two upper branches formed due to the mixture between upper polaritons $|U\rangle \approx (|1X\rangle - |C\rangle)/2$ and dark excitons $|2X\rangle$. We emphasize that if $\delta \neq 0$ both upper polariton eigenstates $|U_-\rangle$ and $|U_+\rangle$ contain fractions of the bright exciton and photon and of the dark exciton. This allows for their direct optical excitation as well as for the radiative transition between the states $|U_\pm\rangle$ and the lower polariton state $|L\rangle$ accompanied by emission of a terahertz photon. The rate of the spontaneous emission of terahertz radiation can be estimated from the Planck formula

$$W_\pm \approx \frac{\omega^3 |d_\pm|^2 n}{3\pi\varepsilon_0 \hbar c^3} F_P = W_0 F_P, \qquad (5)$$

where the dipole matrix element of the optical transition with emission of a terahertz photon in-plane of the cavity is given by

$$d_\pm = e\langle U_\pm | z | L\rangle \approx \frac{16 b_\pm e L_z}{9\pi^2 \sqrt{2(1+b_\pm^2)}}, \qquad (6)$$

where the last equality holds for a QW of width L_z with infinite barriers. Equation (5) has a form of the product of is the probability of terahertz emission in free space W_0, and the Purcell factor F_p, which describes the enhancement of the rate of the spontaneous emission of terahertz photons due to the presence of the terahertz cavity. The principal effect of the cavity is to increase the electric field operator by a factor of $\sqrt{Q}$ within the frequency band $\Delta\omega \sim \omega_0/Q$ around the cavity resonance frequency ω_0, where Q is the quality factor of the cavity.[11] In the case of a narrow spectral width of the electronic oscillator coupled to the cavity, this results in the Purcell formula[10]

$$F_P = \xi Q, \qquad (7)$$

where ξ is a geometric factor inversely proportional to the cavity volume V [$\xi \approx 1$ for V $V \approx (\lambda/2)^3$, where λ is the radiation wavelength]. In our case, the electronic resonance is broadened due to short lifetimes of the initial (UP) and final (LP) states. Applying Fermi's golden rule to a cavity tuned to be in resonance with a transition between homogeneously broadened levels characterized by lifetimes τ_i and τ_i yields the following generalization of the Purcell formula:

$$F_P = \xi [Q^{-1} + (\omega_0 \tau_i)^{-1} + (\omega_0 \tau_f)^{-1}]^{-1}. \qquad (8)$$

One can easily find that, for lifetimes of the polaritonic states of the order of 10^{-11} s, F_P cannot exceed 10^2, however high Q is. This estimate agrees with the experimental results of Ref. 8.

The generation of terahertz emission by a microcavity in the polariton lasing regime is conveniently described by a system of kinetic equations for the upper and lower polariton states and the terahertz mode. We consider the following experimental situation (see Fig. 2): the hybridized upper states are resonantly optically excited. Created in this way, polaritons relax to the lower polariton states, either directly emitting terahertz photons or via a cascade of $k \neq 0$ states of the lower polariton mode (acoustic phonon assisted relaxation). Considering the degenerate states $|U_-\rangle$ and $|U_+\rangle$ as a single upper polariton state $|U\rangle$, as well as all $k \neq 0$ states as a single reservoir,[13] the rate equations read

$$\dot{N}_U = P - (\tau_U^{-1} + \tau_{UR}^{-1}) N_U + W[N_L N (N_U + 1) - N_U (N_L + 1) \times (N+1)], \qquad (9)$$

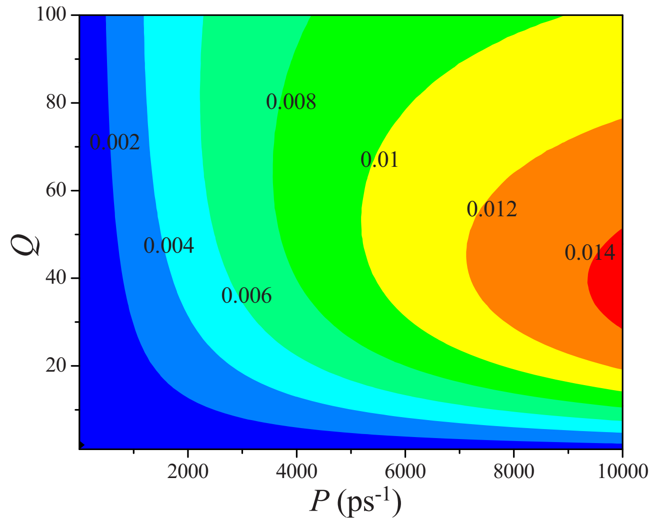

FIG. 3. (Color online) Quantum efficiency of the terahertz emitter as a function of the pump intensity P and a quality factor of the terahertz cavity Q. The black line shows the dependence of the optimum value of Q on pump intensity.

$$\dot{N}_L = -\tau_U^{-1}N_L + \tau_{LR}^{-1}N_R + W[N_U(N_L+1)(N+1) - N_L N(N_U + 1)], \quad (10)$$

$$\dot{N} = -\tau^{-1}N + W[N_U(N_L+1)(N+1) - N_L N(N_U+1)], \quad (11)$$

$$\dot{N}_R = -\tau_R^{-1}N_R - \tau_{LR}^{-1}N_R(N_L+1) + \tau_{UR}^{-1}N_U, \quad (12)$$

where N_U, N_L, N_R, and N are the populations of the upper polariton modes, lower polariton mode, the reservoir of $k \neq 0$ states of the lower polariton mode and the terahertz photon mode, respectively; τ_U, τ_L, τ_R, and τ are the lifetimes of these states, while τ_{UR} and τ_{LR} are the rates of acoustic phonon assisted transitions between the upper polariton mode and the reservoir and between the reservoir and the lowest energy polariton state, respectively. P is the rate of polariton generation in the upper mode due to the optical pump, and W is the rate of terahertz emission given by Eq. (5). In the stationary regime, the occupation number of the terahertz mode can be found from the solution of the above set of equations putting $\dot{N}_U = \dot{N}_L = \dot{N}_R = \dot{N} = 0$. Furthermore, if the terahertz channel of polariton scattering between upper and lower branches would be removed (by setting $W=0$) our system of equations would describe the conventional microcavity polariton system showing the usual features such as polariton lasing characterized by a certain threshold pumping power.[17]

Figure 3 shows the dependence of the quantum efficiency parameter $\beta = N/(\tau P)$ on the pumping rate P and terahertz cavity quality factor Q. The parameters used in this calculation are as follows: $\tau_U = \tau_L = 20$ ps, $\tau_R = 100$, $\tau_R = 100$ ps, $\tau_{UR} = \tau_{LR} = 10$ ps, $\tau/Q = 10$ ps, and $W_0 = 10^{-9}$ ps^{-1}. One can see that for this realistic choice of parameters corresponding to existing polariton lasers,[13,14] the quantum efficiency achieves $\beta = 1.5\%$.

Finally, we note that the mechanism of terahertz emission considered here is qualitatively different from the recently discussed stimulated optical phonon assisted scattering between polariton branches in terahertz cavities.[14] Here we work with conventional optical microcavities in the polariton lasing regime. The terahertz emission is stimulated by the population of the lowest energy exciton-polariton state, no optical phonon assisted relaxation is needed. Semiconductor microcavities in the regime of polariton lasing may be used as efficient sources of the terahertz radiation having a quantum efficiency exceeding 1% according to our estimations.

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