Vertical cavity surface emitting terahertz laser

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Vertical cavity surface emitting terahertz lasers can be realized in conventional semiconductor microcavities with embedded quantum wells in the strong coupling regime. The cavity is to be pumped optically at half the frequency of the 2p exciton state. Once a threshold population of 2p excitons is achieved, a stimulated terahertz transition populates the lower exciton-polariton branch, and the cavity starts emitting laser light both in the optical and terahertz ranges. The lasing threshold is sensitive to the statistics of photons of the pumping light.

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Creation of efficient sources of terahertz (THz) radiation is of high importance for various fields of modern technology, including information transfer, biosensing, security and others. These applications are currently limited due to the lack of compact and reliable solid state sources of THz radiation. The main obstacle preventing the creation of such a source is the low rate of spontaneous emission of THz photons: according to Fermi’s golden rule this rate is proportional to the cube of the frequency and for THz transitions should be roughly tens of inverse milliseconds, while lifetimes of crystal excitations typically lie in the picosecond range. The attempted strategies to improve this ratio include using the Purcell effect by embedding the sample inside a THz cavity, or using the cascade effect in quantum cascade lasers (QCLs). Nevertheless, until now QCLs in the spectral region around 1 THz remain costly, have macroscopic dimensions, are short-lived and still show a quantum efficiency of less than 1%.

Recently, it has been proposed that the emission rate of THz photons may be increased by bosonic stimulation if the THz transition feeds a condensate of exciton-polaritons. In Refs 9, 10 the authors suggest using the transition between the upper and lower polariton branches in a semiconductor microcavity in the polariton lasing regime. The radiative transition between two polariton modes, accompanied by emission of a THz photon, may become allowed if an electric field mixing polariton and dark exciton states is applied to the cavity. THz photons would be emitted in the plane of the cavity, and a supplementary lateral THz cavity would be needed to provide the positive feedback.

Here we propose an alternative model of a microcavity based THz laser, which has significant advantages compared to one based on the transition between two polariton branches. We propose using two-photon pumping of a 2p exciton state, as has been realised already in GaAs based quantum well structures 10, 11. The direct transition to or from the 2p exciton state with emission or absorption of a single photon is forbidden by optical selection rules. Instead, a 2p exciton can radiatively decay to the lower exciton-polariton mode formed by the 1s exciton and cavity photon. This transition is accompanied by emission of a THz photon. The inverse process (THz absorption by a lower polariton mode with excitation of a 2p exciton) has been recently observed experimentally. The THz transition from the 2p state pumps the lowest energy exciton-polariton state, which eventually leads to the polariton lasing effect, widely discussed in the literature. A macroscopic occupation of the lowest energy polariton state stimulates emission of THz photons, so that, in the polariton lasing regime, the cavity would emit one THz photon for each optical photon emitted by the polariton laser, ideally.

The design of the laser and the involved energy levels are illustrated schematically in Figure 1. This design has two crucial advantages with respect to that previously considered: it allows for operation with an optically allowed THz transition, and it provides vertical emission of THz photons. The whole structure is microscopic; no waveguides or THz cavities are needed. This latter point is also a significant advantage with respect to the quantum cascade laser, which operates in the wave-guide geometry. Moreover, the suggested scheme is very interesting from a fundamental point of view, since, as shown below, the threshold of the proposed laser appears to be sensitive to the statistics of photons of the pumping light. Here we present the quantum model of this laser based on the Liouville equation for the density matrix describing the 2p exciton state, the lower polariton mode, and optical and THz photons.

The system under study consists of 2p excitons and 1s
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branches, as well as the 2p exciton state, with frequency \(\omega_p\).

The pump frequency, \(\omega_p\), is half that of the 2p exciton state,

as discussed in the text. (b) The structure considered in this

work: a semiconductor microcavity consisting of an active

layer containing quantum wells (QWs) sandwiched between

two distributed Bragg reflectors (DBRs). The structure is

pumped vertically, i.e., in the direction perpendicular to the

microcavity plane, and the resulting THz emission from the

cavity is in the same direction.

exciton-polaritons interacting with an external electromagnetic field and phonons in a semiconductor microcavity. Hence, our consideration involves both coherent and incoherent processes. Consequently, it is convenient to split the total Hamiltonian of the system into two parts,

\[
H = H_c + H_d,
\]

where the part \(H_c\) includes all types of coherent processes in our system and part \(H_d\) describes the decoherence in the system due to exciton interactions with acoustic phonons, modelled as a classical reservoir\(^{15,16}\).

The equation for the density matrix can be written in the following form:

\[
\frac{d\rho}{dt} = i\hbar [\rho, H_c] + \hat{L} \rho,
\]

where \(\hat{L}\) is a Lindblad superoperator defined below. The incoherent part of the Hamiltonian can be divided into two parts, \(H_d = \sum_i (H_{i+}^c + H_{i-}^c)\), where \(H_{i+}^c\) creates an excitation in the quantum system (and thus annihilates an excitation in the classical reservoir), and \(H_{i-}^c\), conversely, annihilates an excitation in the quantum system and creates an excitation in the classical reservoir. The subscript index \(i\) numerates different types of decoherence processes in the system.

The Lindblad terms can be then written as\(^{16,17}\).

\[
\hat{L} \rho = \frac{\delta(\Delta E)}{\hbar} \sum_i \left\{ (H_{i+}^c \rho H_{i-}^c + H_{i-}^c \rho H_{i+}^c) - (H_{i+}^c H_{i-}^c + H_{i-}^c H_{i+}^c) \rho - \rho (H_{i+}^c H_{i-}^c + H_{i-}^c H_{i+}^c) \right\}, \tag{2}
\]

where \(\delta(\Delta E)\) accounts for energy conservation, and in realistic calculations should be taken as an average inverse broadening of the states in our system, \(\delta(\Delta E) \rightarrow \zeta^{-1}\).

In the particular system which we consider, the coherent and incoherent parts of the Hamiltonian can be written as

\[
H_c = \epsilon_p p^+ p + \epsilon_s s^+ s + \epsilon_a a^+ a + \epsilon_T c^+ c, \tag{3}
\]

\[
H_d = \sum_{i=1,2} (H_{i+}^c + H_{i-}^c), \tag{4}
\]

where

\[
H_{i+}^c = g p^+ a^2, \tag{5}
\]

\[
H_{i-}^c = g p a^2, \tag{6}
\]

\[
H_2^+ = G p^+ sc, \tag{7}
\]

\[
H_2^- = G ps^+ c. \tag{8}
\]

Here \(p\) and \(s\) denote annihilation operators of the 2p exciton and the lowest energy 1s polariton states, respectively, \(a\) is the annihilation operator for laser photons exciting the 2p exciton state, and \(c\) is an annihilation operator for THz photons produced by the 2p \(\rightarrow\) 1s transition. \(\epsilon_p, \epsilon_s, \epsilon_a\) and \(\epsilon_T\) are the energies of the 2p exciton, 1s exciton-polariton, pump photon (2\(\epsilon_a = \epsilon_p\) and THz photon, \(\epsilon_T = \epsilon_p - \epsilon_s\), respectively. Constants \(g\) and \(G\) define the strengths of the two-photon absorption and 1s \(\rightarrow\) 2p radiative transitions, respectively. While the 1s \(\rightarrow\) 2p transition strength is determined by the dipole matrix elements, the main contribution to the two-photon 2p excitation comes from processes with intermediate states in the valence or conduction bands\(^{15}\). We assume that the upper polariton state is higher in energy than the 2p exciton state, and thus can be excluded from consideration. The classical reservoir consists of the photons of the external laser light and THz photons. Note, that the coherent part of our Hamiltonian describes only the free modes of the system and thus is irrelevant to the kinetic equations.

Now, for the occupancy of the 2p exciton mode one can obtain after straightforward algebra:

\[
\frac{dN_p}{dt} = Tr \left\{ p^+ p \frac{d\rho}{dt} \right\} = \frac{2}{\hbar \zeta} \text{Re} \{ Tr \left\{ \rho [H^-; p^+ p; H^+] \right\} \} = W_g \left[ \frac{1}{2} \langle a^+ a^2 \rangle - \langle p^+ p (2 a^+ a + 1) \rangle \right] + W_G \left[ \langle s^+ c^+ c^+ \rangle - \langle p^+ p (s^+ s + c^+ e + c) \rangle \right], \tag{9}
\]

where \(\langle \ldots \rangle = Tr \{ \ldots \rho \} \) denotes averaging with the appropriate density matrix \(\rho\). \(W_g = 4 g^2 / \hbar \zeta \) and \(W_G = 2 G^2 / \hbar \zeta \). The equations for the polariton mode occupancy \(N_s = \langle s^+ s \rangle\), and the terahertz mode occupancy \(N_c = \langle c^+ c \rangle\) are analogous to those for \(N_p\). The occupancy of the pumping mode \(N_p = \langle a^+ a \rangle\) is defined by the intensity of the external pump and we do not need to write an independent dynamic equation for it. The same holds true for the higher order correlators involving pump operators, e.g., \(\langle a^+ a^2 \rangle\). It follows from Eq. 9 that the dynamic equations for the occupancies of the
modes contain quantum correlators of fourth order, such as \((s^+sc^+c)\). For them one can also write the dynamic equations analogous to Eq. (9), which would contain correlators of sixth order. Proceeding further, one would obtain an infinite chain of coupled equations for the hierarchy of correlators.

In order to solve this chain of equations one needs to truncate the correlators at some stage. Here we use the mean-field approximation, which consists in truncation of the fourth-order correlators into products of second-order ones. One can approximate \(\langle p^+pa^+a \rangle \approx \langle p^+p \rangle \langle a^+a \rangle = N_p N_a, (s^+sc^+c) \approx N_s N_c, \ldots\) etc.

A particular analysis is needed for the truncation of the correlator \(\langle a^{+2}a^2 \rangle\) containing four operators corresponding to the pumping mode. Using the definition of the second order coherence \(g^{(2)}(0)\), it can be represented as

\[
\langle a^{+2}a^2 \rangle = g^{(2)}(0)N_a^2. \tag{10}
\]

Finally, the closed set of equations of motion describing the dynamics of our system reads

\[
\frac{dN_p}{dt} = -\frac{N_p}{\tau_p} + W_g \left[ g^{(2)}(0) - N_p(2N_a + 1) \right] + \langle W \rangle G \left\{ N_s N_c(N_p + 1) - N_p(N_a + 1)(N_c + 1) \right\}, \tag{11}
\]

\[
\frac{dN_a}{dt} = -\frac{N_a}{\tau_a} - W_G \left\{ N_s N_c(N_p + 1) - N_p(N_a + 1)(N_c + 1) \right\}, \tag{12}
\]

\[
\frac{dN_c}{dt} = -\frac{N_c}{\tau_c} - W_G \left\{ N_s N_c(N_p + 1) - N_p(N_a + 1)(N_c + 1) \right\}, \tag{13}
\]

where we have introduced the non-radiative lifetime of the 2p exciton state, \(\tau_p\), and lifetimes of lower polaritons and THz photons, \(\tau_a\) and \(\tau_c\), respectively.

Let us analyze in more detail the kinetic equation for the 2p state occupation \(N_p\). The terms describing incoming and outgoing scattering rates for this state can be written as

\[
(1 + N_p)\langle a^{+2}a^2 \rangle - N_p\langle a^{+2}a^2 \rangle.
\]

As we already mentioned in Eq. (10), the incoming rate is proportional to \(g^{(2)}(0)N_a^2\). The outgoing rate can be recast as \(\propto \langle a^{+2}(a^+)^2 \rangle = g^{(2)}(0)N_a^2 + 4N_a + 2\). Hence, all terms containing \(N_a^2 N_p\) cancel each other and Eq. (11) holds. Note, that for processes of non-degenerate two-photon absorption or emission neglected here, two-photon emission processes where photon frequencies are different can be included in \(\tau_p\), the decay rate of the 2p state. The scattering rates have standard form \((1 + N_p)N_a N_a - N_p(N_a + 1)(N_a + 1)\), independent of the photon field statistics. In what follows we assume that \(N_a \gg 1\), making it possible to neglect all other modes.

The key feature of Eq. (11) is that the pumping term \(W_g g^{(2)}(0)N_a^2\) contains the second order coherence of the pump, i.e. strongly depends on its statistics. For a laser pump corresponding to the coherent state this statistics is Poissonian and \(g^{(2)}(0) = 1\). On the other hand different radiation sources may provide different statistics of pump photons and different values of \(g^{(2)}(0)\). For example, for a thermal pump \(g^{(2)}(0) = 2\). Different values of \(g^{(2)}(0)\) lead to different lasing thresholds in the system. Note that dependence of two-photon optical processes on the second order coherence of light has been discussed by Ivchenko in Ref. [23]. In Figure 1, we plot the steady state solutions for \(N_p\) and \(N_s\), assuming that the terahertz mode occupancy \(N_s\) is zero, for the cases of coherent and thermal pumps. For a qualitative understanding, we note that the threshold is reached if \(N_s \sim 1\), or equivalently when \(N_p \sim 1/\tau_p W_G\), since this signals the onset of Bose stimulation of the transition from the 2p-state to the lower polariton, accompanied by terahertz emission. The terahertz generation rate is given by \(T = W_G N_p/N_s + 1 \approx 1/\tau_c N_s\) and therefore shows the same threshold behaviour with pumping intensity as \(N_s\), as can also be seen in Figure 2. We observe that the threshold is higher in the case of coherent pump, which may be understood in terms of the enhanced losses from the 2p-state due to stimulated two-photon emission.

The dynamics of switching the system to polariton and terahertz lasing is essentially governed by the dynamics of the correlator \(\langle a^{+2}a^2 \rangle\), i.e. by the product of squared intensity and second order coherence of the pumping light. The efficiency of two-photon absorption \(W_p\) and the 1s polariton lifetime \(\tau_s\), dependent on the quality factor of the optical cavity, are two main factors which govern the threshold to lasing. The polarisation of pumping defines the polarisation of emission, and the frequency of THz radiation may be tuned by changing the exciton-photon detuning in the microcavity. Interestingly, THz lasing in our system is possible in the absence of a THz cavity.
order to demonstrate this, we have taken the terahertz photon lifetime $\tau_c = 0$ in the calculation shown in Figure 2. One can see that THz lasing begins simultaneously with polariton lasing. The stimulated emission of THz photons is achieved due to the bosonic stimulation of this transition by occupation of the final ($1s$ polariton) state. The system emits THz radiation through the Bragg mirrors of the microcavity, which are transparent at THz frequencies.

In conclusion, we have developed a quantum theory of vertical cavity surface emitting terahertz lasers. The vertical terahertz lasing coexists with polariton lasing and, consequently, is characterised by a low pumping threshold. Interestingly, the value of the threshold is strongly dependent on the statistics of photons of the pumping light. For practical realization of compact terahertz lasers operating at room temperature, microcavities based on wide band gap semiconductors (GaN or ZnO) would seem to be the most advantageous. The resonant two photon pumping of $2p$ exciton states in such a cavity may be assured by a conventional vertical cavity light emitting diode (VCLED) emitting in red. This prospective structure would consist of a GaN or ZnO microcavity grown on the top of a GaAs based VCLED structure. As emission of coherent terahertz light coexists with the polariton lasing in the optical frequency range in this scheme, one can use the visible blue or green light produced by the polariton laser as a marker for the terahertz light beam, which may be important for applications in medicine, security control, etc.

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