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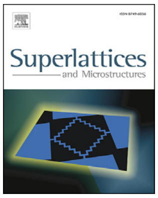

Bosonic cascades of indirect excitons

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ABSTRACT

Recently, the concept of the terahertz bosonic cascade laser (BCL) based on a parabolic quantum well (PQW) embedded in a microcavity was proposed. We refine this proposal by suggesting transitions between indirect exciton (IX) states as a source of terahertz emission. We explicitly propose a structure containing a narrow-square QW and a wide-parabolic QW for the realisation of a bosonic cascade. Advantages of this type of structures are in large dipole matrix elements for terahertz transitions and in long exciton radiative lifetimes which are crucial for realisation of threshold and quantum efficiency BCLs.

1. Introduction

The concept of quantum cascade laser (QCL) was proposed in 1971 by Kazarinov and Suris [1]. They demonstrated that an electromagnetic field may be amplified by a series of consecutive intersubband electron transitions in a semiconductor superlattice in the presence of the electric field applied in the normal to layer plane direction. The high quantum efficiency of the QCL is due to the fact that each optically or electrically excited electron emits several photons while falling down a ladder of equidistant discrete levels. This theoretical scheme was realised more than twenty years later in a semiconductor structure grown by a molecular beam epitaxy [2]. Despite certain frequency tunability, the intersubband transitions usually limit the QCL emission to the infrared frequency range.

Perhaps the most important limiting factor of the QCL is the Pauli exclusion principle, which prevents an electron from subsequent transition to the lower level of the ladder if the latter is occupied. The idea of bosonic cascade laser (BCL) has been recently proposed [3] to evade this constriction of the fermionic statistics. In contrast to the kinetics of fermions, bosonic kinetics yields stimulation of the particle transitions down the cascade ladder by the occupation of the lower levels. In the original proposal an electron undergoing the cascade of transitions was replaced with the exciton, while the essential feature of the quantum cascade, the ladder of equidistant levels, stemmed from the spatial quantization of the excitonic motion in a parabolic quantum well (PQW) potential.

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The possibility of terahertz (THz) lasing in a single PQW was predicted with the use of the semiclassical Boltzmann equation approach [3]. In a later work, second quantization analysis supported this prediction and revealed the superbunching effect in the optical emission of the cascade [4]. The quantum efficiency of THz emission above unity was demonstrated to be feasible without a THz cavity, while the presence of a THz cavity allows for the double stimulation of the THz emission, lowers the pumping threshold of the cascade onset, and leads to the stabilization of the phase of emitted THz photons.

The experimental study of PQWs became possible with the progress on semiconductor structure growth technique, mainly in AlGaAs systems [5]. However, recent study of the exciton dynamics [6,7] in a wide single PQW demonstrated that the exciton energy relaxation does not follow the BCL model of Ref. [3]. In particular, the phonon assisted energy relaxation was shown to be dominant over the radiative cascade relaxation, which implies suppression of the radiative transitions.

In Section 2 we discuss a possible reason of the radiative channel suppression, namely, decoupling of the exciton internal and external degrees of freedom. In Section 3 we propose an alternative scheme of BCL in double QW structures containing indirect exciton (IX) levels with naturally coupled internal electron-hole motion and the center of mass motion. IXs are formed in double quantum wells (DQWs) or electrically biased wide single QWs and consist of an electron and a heavy hole occupying different QWs.

Having the advantage of extremely long lifetimes, governed by vanishingly small electron-hole overlap, IXs in DQWs may be pumped at high densities and macroscopically occupy single quantum states [8].

The density of IX gas is limited approximately by the squared inverse exciton Bohr radius, the limit imposed by the excitonic Mott transition, occurring at [9]. Intersubband electronic transitions are dipole-allowed, however, they are suffiently efficient as multiple studies of QCL show. The transitions between IX states with the same hole are essentially governed by intersubband electronic transitions. As the bosonic stimulation effect is linear in the final state occupation numbers, long-living IX condensates may provide suffiently short THz photon emission times, making the radiative relaxation channel preferential. The double stimulation of THz emission by the IX condensate occupation number and by the THz cavity mode may lead to a high quantum efficiency of BCL based on IXs.

2. Limitation of the single PQW scheme

The basic operation scheme for the BCL proposal of Ref. [3] is sketched Fig. 1, where the different coloured wavefunctions represent different center of mass excitonic states confined in the harmonic potential of a characteristic frequency $\Delta/\hbar$. The horizontal arrows labelled V_1 represent a coupling between the center of mass motion and the internal excitonic degrees of freedom. The diagonal arrows labelled V_d correspond to the dipolar radiative transition $2p\rightarrow 1s$ coupling different internal quantum states of the exciton. Investigating this scheme we can pinpoint a few possible reasons that could hinder observation of the expected THz transitions. On one hand, the limited magnitude of V_1 requires a fine tuning of the inter-level energy distance Δ to the energy of the radiative hydrogen-like transition of the internal electron-hole motion $E_{2p}-E_{1s}$, with even modest shifts due to the imperfection of the PQW leading to drastic reduction of the probability of the exciton radiative relaxation. On the other hand, the theory in Ref. [3] is developed at perfect resonance condition $\Delta = E_{2p}-E_{1s}$, using the linear superpositions between the $1s$ exciton at the n-th size-quantized level and the $2p$ exciton at the level $n-1$ as ladder steps. This treatment is strictly speaking correct only for $V_1 \gg V_d$, and while this condition can be fulfilled at the single-exciton level, it is still unclear if it holds also if a dense gas of excitons is present. Those effects constitute potential bottlenecks of the BCL scheme in a single PQW. Another possible reasons for the discrepancy between the experimental data and the model prediction is the difference in the assumed and practical pumping schemes. Incoherent pumping of the cascade from a hot exciton reservoir was used experimentally, while the initial theoretical proposal assumed the resonant pumping scheme.

Here we propose a modified BCL scheme that allows for a significant improvement of the quantum efficiency of this device in realistic conditions.

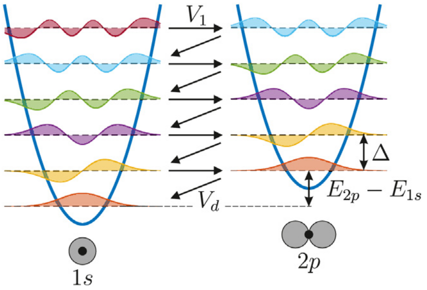

Fig. 1. Single PQW bosonic cascade scheme. Cascade ladders for 1s and 2p state exciton are put in resonance by the fine tuning of internal exciton motion energy with the interlevel energy distance. The cascade route may be then subdivided in two steps, where the THz quantum emission follows from 2p to 1s exciton transition.

3. Double PQW scheme

The undesirable decoupling of the internal electron-hole motion from the exciton center of mass degree of freedom and subsequent suppression of the radiative exciton transitions may be evaded by spatial separation of electron and hole, exploiting a double QW structure shown in Fig. 2a, consisting of a narrow QW and a PQW. The narrow QW on the left of the figure supports a single electron and hole state and plays a role of the reservoir feeding the cascade. Electron-hole pairs can be directly generated in this QW with resonant optical pumping, or, alternatively, can be captured in the QW from the bulk of the structure, where they are generated electrically. The separation between the two QWs may be chosen in a way to allow electron tunneling from the narrow to the wide parabolic QW, while effectively suppressing heavy hole tunneling. The initial feeding of the PQW with electrons tunneling from the narrow QW is accompanied with formation of IXs. Assuming that the exciton binding energy is small compared to the inter-level energy distance, while still retaining the bosonic character of the excitations, the IXs may follow the bosonic cascade route, which is mostly governed by the electron intersubband transition, while the hole remains in the same size-quantized state of the narrow QW. Transition selection rules directly follow from the selection rules of the dipole transitions of the harmonic oscillator: nonzero dipole transition elements only couple neighbouring energy levels, while the absolute value of the matrix element linearly scales with the initial state quantum number. Hence, the first condition for BCL is automatically satisfied, and the only allowed stimulated transition in the dipole approximation is the nearest neighbouring level hopping. Notice that, in first approximation, the effect of electron Coulomb attraction to the hole, which occupies the narrow QW, does not affect the equidistance of the electron subbands: the homogeneous part of the electric field produced by the hole at the PQW only shifts the harmonic potential, not altering its form. Below we present also the calculated IX binding energies for the considered ladder of states.

In our opinion, a very significant advantage of this scheme is that it does not require any fine tuning of the two energies: internal electron-hole excitation energy $|E_{2s} - E_{1s}|$ on one hand, and the size quantization energy of the exciton as a whole in the PQW on the other hand. Moreover the internal and external degrees of freedom for electrons and holes are necessarily coupled due to the spatial separation by the inter-well barrier. This allows resolving potentially overcoming some of the issues highlighted in Section II.

To be specific, we now consider the double QW structure shown in Fig. 2, using GaAs-AlGaAs material platform. We consider an extremely narrow rectangular QW of the width of 2 nm and a wide PQW of the width of 32 nm. The aluminium fraction in the barrier is taken to be $x = 0.33$. Tunneling rates out of the narrow QW may be estimated for the electron, as well as the light and heavy holes, using

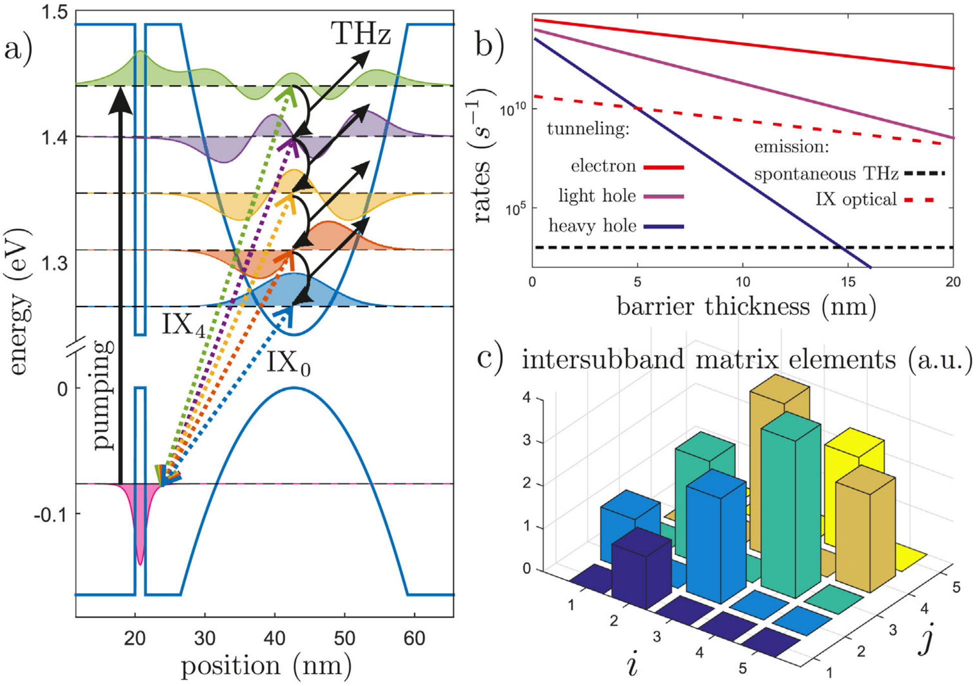

Fig. 2. Double QW bosonic cascade scheme. a) energy diagram of the model AlGaAs-based structure and the single carrier eigenstates. The bosonic cascade is pumped at the direct exciton resonance in the narrow QW. Indirect excitons, formed by an electron in the PQW and a heavy hole in the narrow QW, then follow the bosonic cascade route, emitting a THz quantum at each step. b) Characteristic rates comparison: heavy hole tunneling becomes slower than the spontaneous THz emission only at extremely thick barriers, but is slower then all the other rates of the system for reasonable barriers. c) Transition elements $M_{i,j}$ calculated on the indirect exciton wavefunctions forming the cascade. The nonzero elements only couple the neighbouring level transitions.

$$T_{e(lh,hh)}(d) \sim \frac{E_{e(lh,hh)}}{\hbar} \exp\Big(- \kappa_{e(lh,hh)} d \Big), \tag{1}$$

where $E_{e(lh,hh)}$ refers to free carrier size quantization energy in the narrow QW, $\kappa_{e(lh,hh)} = \sqrt{2m_{e(lh,hh)}(U - E_{e(lh,hh)})}/\hbar$, U is the barrier height, and d is the barrier thickness. We assume that the heavy hole tunneling is slow on the scale of the exciton lifetime. The exciton radiative emission rate may be in turn calculated using [10].

$$\Gamma_{\rm IX}(d) = \frac{4\alpha}{\sqrt{\varepsilon}} \frac{E_{\rm IX}}{\hbar} \left(\frac{|d_{\rm CV}|}{a_B} \right)^2 I^2, \tag{2}$$

where $\alpha \approx 1/137$ is the fine structure constant, $d_{\rm CV}$ is the dipole optical matrix element of the material, a_B is the IX Bohr radius, and $I = \int \Psi_e(z)\Psi_h(z)\mathrm{d}z$ is the electron-hole overlap integral in the growth direction. The latter in turn is governed by the electron tunneling from the PQW back to the narrow QW and is thus proportional to $\exp(-\kappa_e d)$. The results of the performed estimations using Eqs.(1) and (2) are plotted in Fig. 2b. The slowest timescale of the system is the spontaneous THz emission rate. Heavy hole tunneling rate only becomes comparable to it at extremely thick barrier widths. However, for realistic barriers thicker than 5 nm the heavy hole effectively stays in the narrow QW on the timescale of the IX radiative emission, making IX dynamically stable. This is sufficient for the THz emission cascade to switch on: in the regime of stimulated relaxation of excitons down the cascade ladder the THz transition rate is amplified by the number of excitons, and the slowest timescale of the system is given by the IX lifetime. At the same time, the light hole may be taken out of consideration, as any light holes present in the narrow QW freely tunnel through the barrier and die out through the recombination with electrons in the PQW.

Based on this argument, we take the barrier width between the two QWs $d = 5$ nm. The single carrier wavefunctions are plotted along with the energy diagram of the model structure in Fig. 2a. The resonance between the 5th PQW electron subband and the electron level of the narrow QW facilitate the electron tunneling, while the corresponding hole state confined in the narrow QW is effectively decoupled from the hole states localised in the wide PQW.

We use the configurational interaction approach to calculate the excitonic states of the cascade from the single electron and hole states. In the Born-Oppenheimer approximation we separate the electron and hole motion and expand the exciton wavefunction into a linear combination of configurations:

$$\Psi(z_e, z_h, \rho) = \sum_j \Psi_h(z_h)\Psi_e^i(z_e)\Psi^i(\rho), \tag{3}$$

where ρ is the electron-hole distance in the QW plane, and the index i spans over the confined single electron states of the cascades. Following ref. [10], we reduce the Shroedinger equation to coupled equations on the electron-hole relative motion:

$$\left[-\frac{\hbar^2}{2\mu} \frac{1}{\rho} \frac{\partial}{\partial \rho} \rho \frac{\partial}{\partial \rho} + E_e^j + E_h \right] \Psi^i - \frac{e^2}{\varepsilon} \sum_j \Psi^j \int \frac{\Psi_h(z_h)^2 \Psi_e^i(z_e) \Psi_e^j(z_e)}{\sqrt{\rho^2 + (z_e - z_h)^2}} \mathrm{d}z_e \mathrm{d}z_h = E_X^i \Psi^i, \tag{4}$$

where e is the elementary charge, ε is the material electrical permittivity and μ is the reduced exciton mass. The Coulomb attraction matrix elements are of the order of 1 meV, while the interlevel distance of the electron subspace is of the order of 10 meV, allowing us to neglect the mixing of electron states and omit all the terms in the summation, except the one with $i = j$:

$$\left[-\frac{\hbar^2}{2\mu} \frac{1}{\rho} \frac{\partial}{\partial \rho} \rho \frac{\partial}{\partial \rho} + E_e^j + E_h - \frac{e^2}{\varepsilon} \int \frac{\Psi_h(z_h)^2 \Psi_e^i(z_e)^2}{\sqrt{\rho^2 + (z_e - z_h)^2}} \mathrm{d}z_e \mathrm{d}z_h \right] \Psi^i = E_X^i \Psi^i. \tag{5}$$

These equations may be solved with variational method, the trial function being

$$\Psi^i = \sqrt{\frac{2}{\pi}} \frac{1}{a} \exp\Big(-\frac{\rho}{a} \Big), \tag{6}$$

where a is the variational parameter. The kinetic energy term calculated on the trial function (6) yields:

$$E_k = \frac{\hbar^2}{2\mu a^2}. \tag{7}$$

The Coulomb attraction term E_p may be calculated numerically. The calculation of the exciton binding energy for the considered structure yields $E_b \approx 2$ meV, which is one order less than the interlevel distance. The variation of the binding energy from the bottom to the top level of the excitonic cascade δE_b is in turn one order lower the characteristic binding energy $E_b \gg \delta E_b$. We argue that the variation of the exciton binding energy preserves the equidistance of the THz transitions, as it is small compared to the characteristic THz cavity linewidth, given by E_{THz}/Q, where $Q \approx 20$ is the cavity quality factor.

Finally, the interlevel transition dipole elements may also be numerically calculated, taking the exciton wavefunction as the direct product of the single electron and hole wavefunctions. The computed transition elements are plotted in Fig. 2c. Numerical calculation confirms that only the transitions between the neighbouring levels of the cascade are emitting THz photons in the dipole approximation. These transitions are stimulated both by the IX occupation numbers and by the occupation number of the THz photon mode.

4. Discussion

We have presented a proposal for the use of IXs in double quantum well structure to implement a BCL. The proposed scheme relaxes the requirement of the fine energy tuning between the 2*p*-1*s* exciton transition energy and the interlevel energy distance. Moreover, it provides excitons with a strong coupling between internal and center of mass degrees of freedom, that maximizes the matrix elements of THz transitions in the cascade. In addition, long lifetimes and intrinsic dipole moments of IXs help accumulating large numbers of IXs in the structure, which is crucial for triggering the THz transition. A potential drawback of the proposed structure is the lowering of the exciton binding energy, which makes excitons more fragile versus temperature and interaction induced dissociation. For the considered model AlGaAs structure the IX states taking part in the bosonic cascade are characterised by binding energies of the order of 2 meV, limiting the potential device application to extremely low temperatures. However, the importance of this factor can be strongly reduced by choosing the material platforms with strong excitonic effects and high bulk exciton binding energies, e.g. GaN or ZnO.

It should also be noted that the proposed scheme, relying on spatially indirect excitons, does not need an electric field to be operational, while the spatial electron-hole separation is provided by stimulated relaxation of exciton energy in the bosonic cascade. In addition, it allows for both optical and electrical injection of carriers.

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