Superradiance of nearly condensed excitons in InAs-GaSb coupled quantum wells

B. Laikhtman and L. D. Shvartsman
The Racah Institute of Physics, The Hebrew University, Jerusalem 91904, Israel
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We suggest a system of ground state excitons in multiple InAs-GaSb coupled quantum wells as a new source of superradiance. Coupled InAs-GaSb quantum wells can be tuned to a very large wavelength that makes it possible to fabricate a sample with a large number of quantum wells radiating in phase, and in this way to increase the superradiant effect. Depending on the parameters of the system, this superradiance may be a direct analogy of the known Dicke case, or a realization of new type N-level superradiance. In the last case, the time dependence of the luminescence presents N decaying peaks. This dependence may be considered as a manifestation of transition to the Bose condensate phase.

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The possibilities of observing the Bose-Einstein condensation (BEC) of excitons in nanostructures are widely discussed in the literature over the last decade (see, e.g., Refs. 1–3). This task is nontrivial because of the relatively short lifetimes of excitons and the associated difficulties to get a macroscopic amount of excitons in the same quantum state. The indication of BEC phase is usually thought to be the spectral shape of the luminescence line.

In this paper we suggest a different approach. Instead of the actual BEC phase, i.e., a macroscopic amount of excitons in the ground quantum state, we consider a set of identical subsystems, each containing a small number of excitons in the same quantum state. On one hand, this can be considered as a transition to the actual BEC phase and, on the other hand, this situation is easier to realize.

Each subsystem of M excitons in the same quantum state is a system of $M+1$ equidistant levels. The level separation equals the exciton energy $\hbar\omega$ and in this sense a subsystem is equivalent to an oscillator excited to the Mth level. Different subsystems luminesce as different oscillators or emitters, i.e., a single act of luminescence corresponds to the optical transition of a subsystem to the lower adjacent level that is equivalent to recombination of one exciton. The whole system consists of N such $(M+1)$-level emitters or, in other words, each of these N emitters contains M excitons.[4] (The terms "oscillator" and "emitter" hereafter have the same meaning.) The maximal number of excitons in the whole system is NM, i.e., the total number of levels is $NM+1$. All levels, except the lowest and the highest ones, are degenerate.

As an indication of the transition process to the BEC phase we suggest looking for the nontrivial kinetics of photoluminescence. This system should exhibit superradiance phenomenon that results in a strongly nonmonotonous time dependence of photoluminescence kinetics.[5] For instance, we show that decay of photoluminescence (PL) of certain InAs-GaSb coupled quantum wells (CQWs) having five ground state excitons per system may demonstrate up to five consecutive maximums followed by exponential decay.

In what follows, we first consider a system of N identical emitters (oscillators) interacting with electromagnetic field and then formulate conditions of the realization of these emitters as a system of nearly condensed excitons in quantum wells. The size of the volume containing the emitters, L, is assumed to be much less that the radiation wavelength λ

$$L \ll \lambda. \tag{1}$$

Then the Hamiltonian of N emitters (oscillators) and the field is

$$H = \hbar\omega \sum_{j=1}^{N} a_j^\dagger a_j + \sum_{\nu\mathbf{q}} \hbar c q b_{\nu\mathbf{q}}^\dagger b_{\nu\mathbf{q}} + \sum_{\nu\mathbf{q}} \frac{e\mathbf{p}\mathbf{e}_{\nu\mathbf{q}}}{m_0} \psi_X(0) \sqrt{\frac{2\pi\hbar n_r}{cq\varepsilon V}} \left(b_{\nu\mathbf{q}}^\dagger \sum_{j=1}^{N} a_j + b_{\nu\mathbf{q}} \sum_{j=1}^{N} a_j^\dagger \right), \tag{2}$$

where $\mathbf{p}$ is the momentum matrix element, $\mathbf{e}_{\nu\mathbf{q}}$ is the polarization vector of a photon with wave vector $\mathbf{q}$, and polarization ν, m_0 is the free electron mass, ε is the dielectric constant, n_r is the refraction index, $\psi_X(\mathbf{r})$ is the exciton wave function, $b_{\nu\mathbf{q}}$ and $b_{\nu\mathbf{q}}^\dagger$ are the operators of photon annihilation and creation, and a_j and $a_j^\dagger$ are the operators of exciton annihilation and creation in the jth oscillator. Excitons are assumed to be localized (see the discussion about the realization of the system below) and no momentum conservation is required.

An important feature of the Hamiltonian is that the interaction of the electromagnetic field with the emitters is symmetric with respect to all emitters. Therefore, the interaction of the emitter ensemble with the radiation does not change the symmetry of the ensemble wave function. In particular, the ground state of the ensemble is symmetric with respect to all emitters (all emitters are at the ground state) and any excited state produced with the help of resonant radiation also has to be symmetric with respect to all emitters. Apparently, luminescence also keeps this symmetry.

Below, we point out that except for resonant optical excitation, there can be other ways to obtain symmetric excited states of the exciton ensemble. At the moment we want to stress that if the initial ensemble wave function is symmetric with respect to all emitters, the interaction with radiation does not violate this symmetry. In this respect the emitter

ensemble resembles an ensemble of bosons with internal degrees of freedom. The key difference between the emitter ensemble and the ensemble of bosons is that, in general, there exist ways to bring the former ensemble to a state with any symmetry (e.g., it is possible to excite only some of the emitters making use of their different spatcial location), but these ways are different from the equal interaction of all emitters with the resonant radiation ($L \ll \lambda$). We will discard these ways and consider only symmetric states of the ensemble.

In symmetric states, different emitters are indistinguishable and each state is completely characterized by the set of numbers $k_0, k_1, k_2, \ldots$ that indicates how many emitters are at the ground state (k_0), at the first excited level (k_1), at the second excited level (k_2), and so on. The wave function of an ensemble of N emitters with these quantum numbers is

$$\Psi_N(k_0,k_1,\ldots) = \sqrt{\frac{k_0!k_1!k_2!\cdots}{N!}} \times \sum_{P(i)} \psi_{i_1}(0)\psi_{i_2}(0)\cdots\psi_{i_{k_0}}(0)\psi_{i_{k_0+1}} \times(1)\psi_{i_{k_0+2}}(1)\cdots\psi_{i_{k_0+k_1}}(1)\cdots, \quad (3)$$

where $\psi_i(l)$ is the wave function of the ith emitter at the lth excited state and the summation is made over all permutations of the subscripts i. The probabilities of the transition from state $(k_0,k_1,k_2,\ldots,k_{l-1},k_l,k_{l+1},\ldots)$ to state $(k_0,k_1,k_2,\ldots,k_{l-1}+1,k_l-1,k_{l+1},\ldots)$ with emission of a photon and to state $(k_0,k_1,k_2,\ldots,k_{l-1},k_l-1,k_{l+1}+1,\ldots)$ with absorbtion of a photon calculated with the help of functions (3) are

$$W^{k_0,k_1,k_2,\ldots,k_{l-1}+1,k_l-1,\cdots}_{k_0,k_1,k_2,\ldots,k_{l-1},k_l,\cdots} = \frac{l(k_{l-1}+1)k_l}{\tau}, \quad (4a)$$

$$W^{k_0,k_1,k_2,\ldots,k_l-1,k_{l+1}+1,\cdots}_{k_0,k_1,k_2,\ldots,k_l,k_{l+1},\cdots} = \frac{(l+1)k_l(k_{l+1}+1)}{\tau}, \quad (4b)$$

where $\tau=\hbar\varepsilon(c/n_r)^3m_0^2/2\omega e^2p^2$ is the single exciton lifetime.[6] Factors l and $l+1$ in Eq. (3) are the usual Bose factors. The factors, k_l, $k_{l-1}+1$, and $k_{l+1}+1$ have precisely the same origin. Namely, the emitters in the symmetric states are indistinguishable, and therefore, the probability of an increase in the number of emitters at level l is proportional to k_l+1, while the probability to a decrease in the number of emitters at this level is proportional to k_l.

It is possible to write down an equation describing the dynamics of the average occupation numbers k_l in the course of the radiation process. During time interval dt, occupation number k_l decreases due to photon emission by an emitter at level l, with the probability $W^{k_0,k_1,k_2,\ldots k_{l-1}+1,k_l-1,k_{l+1},\cdots}_{k_0,k_1,k_2,\ldots k_{l-1},k_l,k_{l+1},\ldots}$, and increases due to photon emission by an emitter at the upper level $l+1$ with the probability $W^{k_0,k_1,k_2,\ldots,k_{l-1},k_l+1,k_{l+1}-1,\ldots}_{k_0,k_1,k_2,\ldots,k_{l-1},k_l,k_{l+1},\ldots}$. These probabilities are given by the Eq. (4a). The resulting equation is

$$\frac{dk_l}{dt} = \frac{1}{\tau}[(l+1)k_{l+1}(k_l+1) - lk_l(k_{l-1}+1)]. \quad (5)$$

In a sense, the derivation of this equation is similar to the derivation of a regular Boltzmann equation. For the particular case of superradiance of a two-level system, i.e., the Dicke model, this equation has been obtained by Eberly and Rehler.[7,8] Equation (5) is an approximation, where discreet values of k_l are replaced with continuous values. Strictly speaking this approximation is valid when $k_l \gg 1$. An obvious integral of motion of Eq. (5) is the total number of emitters $N=\Sigma_l k_l$.

The luminescence of the ensemble is characterized by a radiation rate that is a measure of the decrease of the number of excitations (in our case the rate of exciton recombination),

$$I = -\hbar\omega\frac{dn}{dt} = \frac{\hbar\omega}{\tau}\sum_{l=1}^{\infty} lk_l(k_{l-1}+1). \quad (6)$$

Here

$$n = \sum_{l=1}^{\infty} lk_l \quad (7)$$

is the total number of excitations (excitons) in the system. In reality the summation in Eqs. (6) and (7) is limited by the maximal excited level of the emitters. If each emitter can be excited up to the Mth level, the maximal number of the excitations in the system is NM. The energy spectrum of the emitter system is a set of $NM+1$ equidistant levels, and the energy of the system is running from 0 to $NM\hbar\omega$.

The character of the radiation crucially depends on the initial state of the system. In a widely known case of two-level emitters, i.e., $M=1$, if the initial state corresponds to $E_{max}=N\hbar\omega$, the resulting radiation kinetics exhibits one peak followed by an exponential decay, where a typical time scale of the decay is proportional to N^{-1} and the maximal radiation intensity is scaled as N^2 (Ref. 9). The qualitative picture of the peak formation is connected to a specific form of the radiation matrix element. Qualitatively, this form follows from the fact that an intermediate state of the total energy $E_{max}/2=N\hbar\omega/2$ can be realized by the maximal number of microstates $N!/(N/2)!^2$. If one chooses $E<E_{max}/2$ as the initial state, there is no peak in the radiation kinetics.

In the case considered here, $M>1$, we deal with the nonobvious generalization of ordinary superradiance, and the situation becomes much richer. If one now chooses the initial state corresponding to $E_{max}=NM\hbar\omega$, the resulting radiation kinetics exhibits a series of peaks followed by an exponential decay, as shown in the Fig. 1. The number of peaks equals M, but every next peak is a bit more eroded than the previous one.

The physical reason for such a behavior can be easily understood. At the beginning of the process the pair of Mth and $(M-1)$th levels can be considered as the Dicke two-level system and the radiation reaches its maximum when approximately one-half of the emitters are already at the $(M-1)$th level. Then, the appearance of the first peak is a manifestation of ordinary two-level superradiance. The length of the

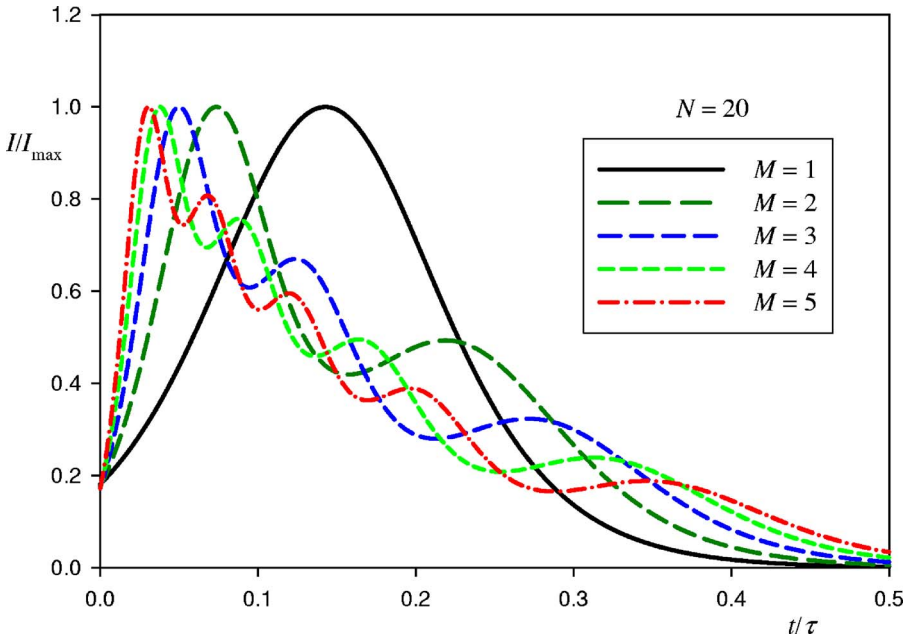

FIG. 1. (Color online) Time dependence of the normalized radiation rate of equally excited 20 emitters. If initially the emitters are excited to the first level (1 exciton in each emitter), the radiation rate has 1 peak. Excitation to the second level (2 excitons in each emitter) gives 2 peaks, excitation to the third level (3 excitons) gives 3 peaks, and so on. The peaks are followed by exponential decay.

peak is roughly proportional to $1/N$, while the intensity is proportional to N^2. Indeed, in the Fig. 2 one can see that for the same number of levels M, the peaks are more pronounced for a larger number of emitters N. The higher number M is, the weaker these dependencies of peak lengths and intensities on N become, because the small corrections associated with other levels become more substantial. At the next stage, after the most of the emitters have passed to the $(M-1)$th level, the radiation related to transition between the pair of $(M-1)$th and $(M-2)$ levels becomes significant. These levels can be considered as another Dicke two-level system and its radiation reaches the maximum when the levels of the next lower pair are equally occupied. The next maximum corresponds to the transitions between the $(M-2)$th and $(M-3)$th levels, and so on.

This behavior can be easily illustrated mathematically. We start with the standard picture of superradiance, i.e., from the two-level system, where $M=1$, and Eq. (5) can be solved analytically,[7,8]

$$k_1(t) = \frac{N+1}{e^{(N+1)(t-t_1)/\tau}+1}, \quad t_1 = \frac{\tau}{N+1}\ln\frac{k_1(0)}{N-k_1(0)+1}, \tag{8a}$$

$$I = \frac{\hbar\omega}{\tau}\frac{(N+1)^2}{4\sinh^2\dfrac{(N+1)(t-t_1)}{2\tau}}. \tag{8b}$$

$I(t)$ has a peak at $t=t_1$ when the number of excited systems is $k_1(t_1)=k_0(t_1)+1=(N+1)/2$, and the peak is followed by the exponential decay, $I(t)\sim(\hbar\omega/\tau)(N+1)^2e^{(N+1)(t_1-t)/\tau}$. The

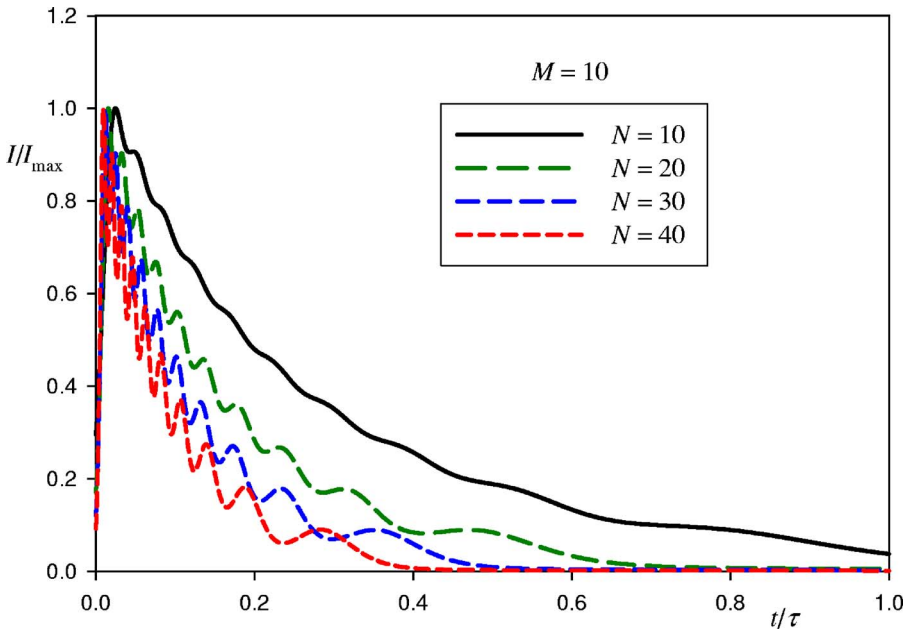

FIG. 2. (Color online) Time dependence of the normalized radiation rate of emitters excited to the 10th level (10 excitons in each emitter). With an increase in the number of emitters, the peaks become more narrow and more pronounced.

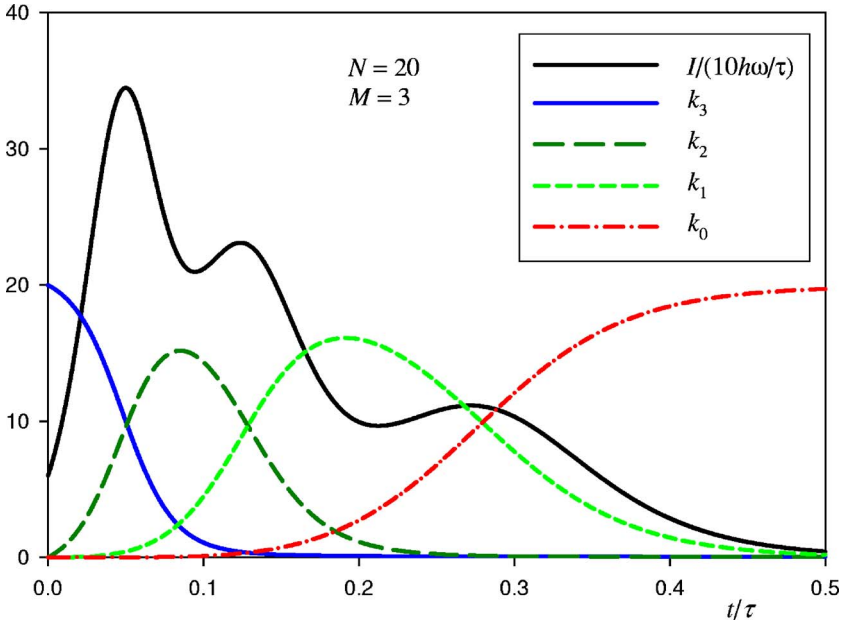

FIG. 3. (Color online) Dynamics of radiation and population of oscillator levels. Peaks of the radiation rate coincide with the equal population of the two neighboring levels.

width of the peak and the exponential decay have the same characteristic time, $\tau/(N+1)$, decreasing with the number of the emitters. The possibility of observing the peak crucially depends on the choice of the initial state. If instead of $k_1(0)=N$, $k_0(0)=0$, one chooses a more smeared population of the initial state, $k_1(0)<(N+1)/2$, the time evolution of the intensity does not exhibit the peak.

In the more general case when $M>1$ the analysis does not change much. If we start from the initial state of fully excited system $k_M=N$, $k_0=k_1=\cdots k_{M-1}=0$, two equations with $l=M,M-1$ can be easily reduced to the above case of a two-level system, at least at the initial stage. Thus, the first peak is formed more or less as for the two-level system. The term with $l=M$ dominates the sum in Eq. (6). There are two basic factors favoring the distribution of the majority of emitters between the maximum two neighboring levels at any time. The first one is the quadratic dependence of the population rate dk_l/dt on k in the right-hand side (rhs) of Eq. (5), which makes transition faster when most of the emitters are equally distributed between two adjacent levels. The second one is the factors $(l+1)$ and l in the matrix elements of optical transition show that transitions happen faster between higher levels. Both of these factors lead to the fact that the existence of the additional levels below is only a small correction that always works as a smearing factor for the ordinary two-level case. This may be seen from the Fig. 3, where we showed the simulated dynamics of the system of $N=20$, four-level $(M=3)$ emitters.[10] The position of each of the three peaks coincides with the instant of nearly equal populations of the corresponding couple of levels responsible for the appearance of these peaks. The first peak corresponds to the optical transition between the levels 4 and 3, and coincides with the point where $k_4(t)=k_3(t)+1\approx(N+1)/2$, the second peak corresponds to the optical transition between the levels 3 and 2 to the point where $k_3(t)=k_2(t)+1\approx(N+1)/2$, etc. Each following peak is wider than the previous one, because for every couple of levels involved, the "initial state" becomes more and more smeared.

The smearing grows with increase of the number of levels M. When M is very large, the radiation originating from the levels different from the major radiating pair becomes more and more important. In the limiting case of very large M, the radiation peaks disappear (see, Figs. 1–3), and the radiation kinetics becomes close to a classical picture of the simple exponential decay without any oscillations.[8]

The smearing, as we show later on, also has very important consequences for the conditions of the experimental observation of the multipeak kinetics discussed. The unavoidable smearing of the initial state makes peaks less pronounced and harder to observe. Because of its principal importance, this phenomena is illustrated in the Fig. 4, where we have chosen the initial state of $N=20$, five-level $(M=4)$ emitters as a Gaussian $k_l(0)=A\exp[-(l-M)^2/2(\Delta M)^2]$ with various degree of smearing. When $\Delta M=1.2$ is reached, the system hardly demonstrates multipeak kinetics.

Returning to well resolved peaks, we can mathematically illustrate the lower intensity of each following peak. According to the picture above, at the maxima of the radiation, the main contribution to the rhs of Eq. (6) comes from only one term, because mainly one couple of levels is populated. The intensity at the maximum corresponding to the transitions between the levels $(l+1)$ and l roughly corresponds to the situation when $k_{l+1}=k_l+1=N/2$, and is proportional to $l(N+1)^2/4$ (smearing of the maxima is neglected in this estimate). During the process of the radiation l corresponding to different maxima becomes smaller that reveals itself in the decay of the envelope of the maxima in time, Figs. 2–4.

We suggest searching for the experimental realization of above phenomena in nearly condensed excitons in quantum wells. The most promising system for this purpose is InAs-GaSb coupled quantum wells (CQWs). This system has a number of potential advantages. Indeed, in general, an experimental observation of the Dicke superradiance on quantum well excitons is problematic. The reason is that usually the radiation wavelength, λ, is much smaller than the typical size of the system, L, and dephasing kills or strongly modi-

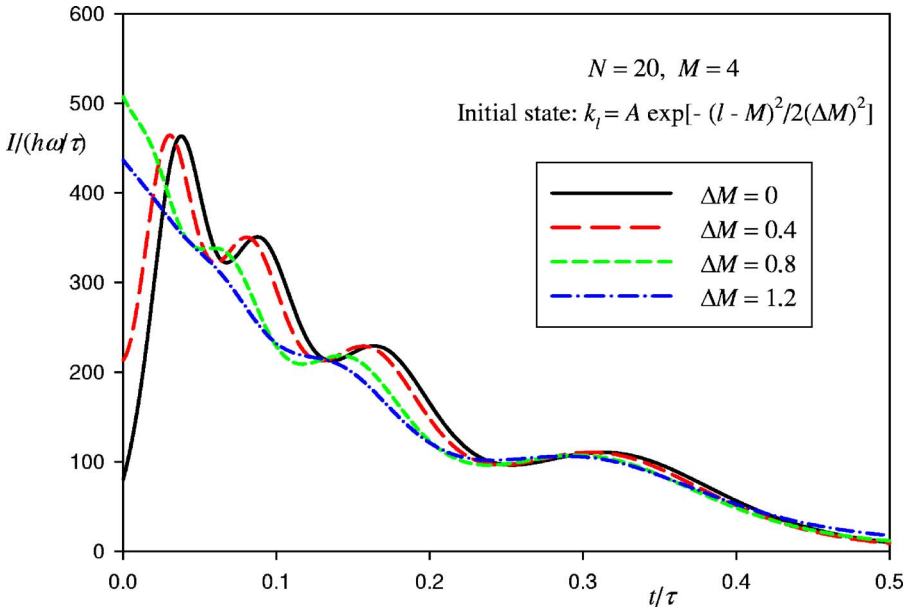

FIG. 4. (Color online) Smearing of the initial level population leads to the smearing of the radiation peaks.

fies the effect.[8] For InAs-GaSb CQWs excitons, the wavelength is tunable, and this ratio may be the opposite, $\lambda \gg L$. In this sense, the situation is close to Dicke. Another advantage of this system is that, as we show below, the exciton lifetime can be made very large.

However, the observation of the multipeak superradiance is a nontrivial problem mainly because the superradiance kinetics is very sensitive to the initial state of the emitter ensemble. We show below that the system of nearly condensed excitons in InAs-GaSb CQWs, with certain growth inhomogeneities excited in a particular way, are equivalent emitters in the same initial quantum state.

It is well known that surface roughness splits exciton levels. Excitons in the ground state are confined in the most wide regions of the well, and each island confining excitons can be considered as a separate emitter. The number of excitons confined in such an island is limited by the relation between the area of the island and the exciton radius. If the average lateral length scale of the surface roughness is W, and the exciton radius (the rms of the exciton wave function radius) is r_0, then the maximal number of excitons that can be confined in one island is W^2/r_0^2. Actually, this number can be smaller because excitons in InAs-GaSb double quantum wells have a dipole moment directed from GaSb to InAs, and the dipole-dipole repulsion prevents confinement of too many excitons in the same island. Thus, the fabrication of wide islands of alike sizes W provides an equal capacity of our emitters.

We consider two ways to create islands of the same size. One is to fabricate a metallic mask on the top of the structure with equal windows (compare Refs. 3 and 11). If the InAs layer is closer to the mask than GaSb layer, then a negative potential at the mask will force excitons to be confined only under the windows. If the GaSb layer is closer to the mask, the same can be done with a positive potential. Also, the windows define areas where excitons can be optically pumped. To eliminate the splitting of the exciton spectrum due to surface roughness in the islands formed by the windows, it is necessary to make the size of each window smaller than the lateral scale of the surface roughness.

The other way is to make use of the natural islands formed by the surface roughness. The area of the islands fluctuates due to fluctuations of their lateral size. The islands contain the same number of excitons if the area fluctuations are not larger than the area occupied by one exciton. If there is no correlation between surface roughness steps, then they are separated on average, by $N_0 \approx W/a_0$ unite cells (a_0 is the lattice constant), and the fluctuation of this separation is $\sqrt{N_0}$. The fluctuation of the island area is $2Wa_0\sqrt{N_0} \approx 2W^{3/2}a_0^{1/2}$. That the lateral size has to meet the condition $2W^{3/2}a_0^{1/2} \lesssim r_0^2$. This means that the number of excitons, N_X, in one island is limited, $N_X \leqslant (r_0/4a_0)^{2/3}$. As a result, the maximal number of luminescence peaks that can be realized in this experimental system is also limited, $M = N_X$.

As an example, we make some estimates for a sample where both InAs and GaSb quantum wells have widths $L_h = L_e = 60$ Å. The exciton radius in such a system[6] is $r_0 \approx 312$ Å, the exciton binding energy is 4.12 meV, the exciton optical energy is 118 meV, and the exciton lifetime is 66 ps. The lattice constant of InAs and GaSb, a_0, is around 6 Å, and this corresponds to no more than five excitons in one island and the roughness lateral scale $W \approx 700$ Å. Therefore, the maximal number of peaks also should not exceed five.

We can also suggest a way to pump equal number of excitons in each island. The excitons can be resonantly pumped.[12–14] In Refs. 12 and 13, exciton concentration $n = 5 \times 10^{10}$ cm^{-2} have been reported for GaAs QWs. In our case of InAs-GaSb CQWs, exciton lifetime is much longer, which facilitates reaching higher densities. The temperature limitation necessary to have all of these excitons in an island in the ground state is not critical in such a case. The temperature has to be smaller than[11]

$$T_c = \frac{2\pi\hbar^2 n}{m \ln(1 + N_X)}. \quad (9)$$

If $n \approx 10^{11}$ (there may be a numerical factor of the order of unity depending on the shape of the widening) and the exciton effective mass $m \approx 0.1$, then $T_c \approx 55$ K.

Exciton lifetime in InAs-GaSb CQWs can be increased even more by the growing of a thin AlSb layer in between the wells. The main recombination channel in this system is electron tunneling from InAs into GaSb, where it recombines with hole[6] (see also Ref. 15). The effective electron mass in AlSb is 0.14, the lattice constant is 6.13 Å, the conduction band offset at the interface InAs/AlSb is 2.07 eV.[16] The layer of one lattice constant width suppresses the electron tunneling by 5.5 times, i.e., makes a lifetime of around 360 ps that is longer than typical energy relaxation time. At the same time, such a thin layer practically does not affect other parameters of the indirect excitons.

In the above estimates we did not take into account dipole-dipole interaction between excitons. The dipole moment of an exciton is $e(L_e+L_h)/2$, and at the distance between the excitons $2r_0$, the dipole-dipole repulsion energy between them is $e^2(L_e+L_h)^2/4\kappa(2r_0)^3 \approx 1.4\times10^{-2}$ meV (here $\kappa \approx 15$ is the dielectric constant). In the structure without an AlSb layer in between InAs and GaSb quantum wells, this energy is of the order of the energy uncertainty due to recombination, 10^{-2} meV, and can be discarded.

To conclude, the luminescence dynamics similar to those shown in the Figs. 1–4 may be realized in resonantly pumped InAs-GaSb CQWs of the above design. Typical τ in this case has to be around 60 ps.

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