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Enhancement and Inhibition of Spontaneous Emission in Room-Temperature Semiconductor Microcavities

Abstract

We have fabricated planar semiconductor microcavities with metallic mirrors in which we have observed both enhancement and inhibition of spontaneous emission, at room temperature. Inhibition is an unambiguous signature of Cavity Quantum Electrodynamics effects, and its observation in these room-temperature semiconductor structures opens the way to novel devices with controlled spontaneous emission.

1 Introduction

Spontaneous emission of light is indispensable in the operation of optoelectronic devices: in Light Emitting Diodes (LED) it constitutes the useful output of the devices, while in semiconductor lasers it provides the first photon that triggers stimulated emission. However, as this phenomenon takes place generally in an uncontrolled way, a very large fraction of the emitted the light is lost. As a consequence, the energy efficiency of LED's is only of the order of a few percent, whereas in semiconductor lasers only 1 photon in 10^5 goes into the lasing mode while all other photons are lost and contribute to raising the threshold of operation of the laser, limiting its bandwidth and introducing noise. Clearly, if the directivity and the dynamics of spontaneous emission can be controlled, this may contribute to greatly increase the efficiency of light emitting diodes, reduce the threshold of lasers, possibly to the point of canceling it, or may lead to the realization of novel light sources with non-classical properties such as sources of controlled trains of single photons.

2 Cavity Quantum Electrodynamics in atoms

Over the last two decades, use of atomic systems, placed in optical cavities of dimensions of the order of the wavelength (called microcavities), has led to the demonstration of the possibility of controlling spontaneous emission, and gave rise to the field of Cavity Quantum Electrodynamics (CQED) [1]. In order to understand how spontaneous emission can be modified inside a cavity, one has to consider simply that this phenomenon is due to the coupling of the excited atomic states to the electromagnetic modes available. Thus, the different modal structure of the electromagnetic field in the microcavity as compared with that in free space produces a modification in the characteristics of spontaneous emission when an emitter is introduced inside such a microcavity.

Two situations are of particular interest. The first one concerns cavities that display very sharp resonances, so that their modal structure consists of a few discrete states. If an atom inside such a cavity couples to a single one of these states (a situation called the "strong coupling" regime) its spontaneous emission will involve a periodic exchange of the energy between the atom and the cavity mode (Rabi oscillations) and, in the spectrum of the light emerging from the cavity, this will give rise to a doublet (vacuum Rabi splitting), rather than to a single emission line at the atomic frequency.

The second situation concerns cavities in which the modal structure constitutes a continuum, such as planar microcavities with ideal metallic mirrors. In this situation, the atom will couple (in what is called the "weak coupling" regime), to the part of the continuum that is at the same frequency as the atomic transition and will undergo a radiative decay of its energy in favor of the electromagnetic modes of the cavity. Near the resonance frequencies of the cavity, the density of states in this continuum is higher than in free space, and this produces an enhancement of the spontaneous emission rate with respect to its value in free space. Below the cut-off frequency of the cavity, on the other hand, the density of electromagnetic states seen by a dipole parallel to the two mirrors falls to zero, implying that in this case there is no coupling between the dipole and the electromagnetic field in the cavity, and thus spontaneous emission is inhibited.

As the development of CQED has been based on considerations and experiments on single atoms that are subject only to the radiative interaction and are otherwise isolated from their environment, it is quite difficult to extend the principles of this theory to semiconductors. Semiconductors are complex material systems that possess very few of the simplifying features of isolated single atoms: In contrast to single atoms, semiconductors involve a very large number of atoms, their electronic excitations present a collective and delocalized character and are subject to numerous interactions with their environment that produce rapid processes of dephasing or energy relaxation.

3 Strong coupling in semiconductors

A major breakthrough in the direction of adapting CQED to semiconductors was accomplished in 1992, when the vacuum Rabi splitting was observed in planar microcavities with high reflectivity Bragg mirrors, containing semiconductor quantum wells, at low temperature [2]. Soon afterwards, it was demonstrated that the spontaneous emission lifetime of the excitons inside the cavity was strongly modified, in accordance with the "strong coupling" theory of CQED [3]. Two features of solid-state physics permit this complex system to meet some of the requirements of atomic CQED. The first one is the translational invariance of the planar cavity that introduces a wavevector selection rule according to which each quantum well exciton can couple to a single mode of the planar cavity, reproducing thus the state-to-state interaction conditions of the atomic "strong coupling" regime. The second one is the low temperature experimental conditions under which the exciton scattering by the thermal lattice vibrations is relatively slow, so that excitons retain essentially the same wavevector and thus satisfy the state-to-state interaction conditions throughout the experimental observation time. On the other hand, when the temperature is raised and the excitons to are rapidly scattered over all wavevectors, the "strong coupling" conditions are not met anymore.

In that case, the spontaneous emission lifetime of the excitons recovers its free space value [3], while the Rabi doublet in the spectrum need not be attributed to CQED, but rather to the classical refractive index variations of the spacer in the vicinity of the exciton frequency [4].

4 Weak coupling in semiconductors

Even though the thermal scattering of carriers in semiconductors compromises the modification of spontaneous emission in the "strong coupling" regime, surprisingly, it can be exploited to meet some of the conditions that permit such a modification under "weak coupling". To understand this, one has to consider that the scattering of carriers and the statistical occupation of a given region in wavevector space can be thought, in direct space, as corresponding to a localization of the extended wavefunctions of the carriers into a statistical distribution of small "coherence volumes" whose extent is given by the Fourier transform of the occupied region in wavevector space. The excited electronic states all of the lattice sites contained in each "coherence volume" are in phase and therefore emit cooperatively as a single dipole, with a spontaneous emission lifetime that is inversely proportional to the number of sites contained in the "coherence volume". At room temperature, the spontaneous emission lifetime in GaAs or InGaAs quantum wells at moderate carrier injection densities (10^{17} cm^{-3}) is of the order of 10 ns, a lifetime that corresponds to a collective oscillator strength of approximately 6 x 10^5 unit cells. This implies that, at room temperature, the elementary collective emitters have a diameter of the order of 200 Angstroms and thus can be considered to be point-like when compared with the wavelength of light emitted by the semiconductor. As a consequence, at room temperature, the CQED behavior of the spontaneous emission of a semiconductor quantum well inside a cavity will be analogous to that of a collection of randomly placed point emitters.

In considering the modification of spontaneous emission of a room temperature quantum well let us first examine the case of a planar cavity bounded by GaAs/AlAs Bragg mirrors. These mirrors present a high reflectivity for angles less than 20 degrees with respect to the normal, while for larger angles (which represent 95 percent of space) the reflectivity drops practically to zero. As a consequence, the spontaneous emission of a point emitter or a room temperature quantum well placed in such a cavity will not be modified appreciably and will retain essentially the same dynamics as in free space [5]. A different behavior is expected, however, for a point emitter placed in a cavity with metallic mirrors where a modification of the dynamics of spontaneous emission is possible. Indeed, metallic mirrors generally present a relatively constant reflectivity over all angles, even if its value is lower than that of Bragg mirrors, at normal incidence, because of dissipative losses in the metal of the order of a few percent. In particular, for GaAs cavities bounded by silver mirrors, preliminary calculations indicate that the spontaneous emission of a dipole oriented parallel to the mirrors should be enhanced by a factor of 4 if the spacer thickness is such that the cavity is resonant with the emitter, whereas for shorter cavities the spontaneous emission of a parallel dipole should be partially inhibited. Because the dissipative losses in the metal introduce additional channels of de-excitation for the dipole, inhibition can never the complete. For the case of silver, which is the metal with the smallest losses, inhibition is manifested by a reduction of the spontaneous emission rate by a factor of 5 [5].

In general, when absorption losses increase, inhibition becomes weaker. Thus, the observation of this phenomenon requires a very careful control of all loss mechanisms. On the other hand, the observation of enhancement is more tolerant to radiative losses and persists even in the presence of absorption in the mirrors or lateral leaks in the cavity. Indeed, for modes that display a very high quality factor and a small volume, the emission rate may be larger than for all other directions of free space, including the leaks, thus permitting a direct observation of enhancement [6]. Under the same conditions, however, where the cavity presents substantial leaks, no inhibition can be observed. Enhancement due to the Purcell effect was recently observed at low temperatures in laterally confined semiconductor microcavities with Bragg mirrors [7]. It should be noted that, in addition to the radiative losses discussed above, there exist a large number of parasitic processes in semiconductors (in particular, non-radiative electron-hole recombination on defects or surface states) that can open fast decay channels for the electron-hole pairs thus masking any enhancement of the spontaneous emission decay rate due to CQED effects. The importance of observing inhibition of spontaneous emission as an unambiguous signature of CQED effects as well as the experimental difficulties associated with this observation were recognized [8] as early as 1981. Inhibition was first observed in Rydberg atoms [9] and was later observed at optical frequencies [10, 11], also in atoms. Until recently, it had never been observed in room temperature solid-state microcavities.

5 Experimental

With the aim of observing inhibition and enhancement of spontaneous emission in room temperature semiconductors, we carried out recently a series of experiments on the photoluminescence dynamics of InGaAs quantum wells placed in GaAs microcavities, bounded by silver mirrors. We fabricated 4 microcavities having spacers of different thickness: (a) 78 nm, (b) 90 nm, (c) 96 nm and (d) 102 nm, each expected to exhibit enhancement or inhibition. A schematic of the microcavity structure is given in Fig. 1.

The spacer of each cavity is an MOCVD-grown multi-layer structure containing in its center a 10 nm $In_{0.13}Ga_{0.87}As$ strained-layer quantum well, emitting at 950 nm at room temperature. The choice of a strained-layer quantum well is important because of the large splitting (40 meV) between the heavy and light hole bands. Because of this splitting, the emission at room temperature involves predominantly the heavy-hole to conduction band transition which is polarized parallel to the quantum well and the mirrors. A 20 nm $Al_{0.40}Ga_{0.60}As$ barrier is grown on either side of the quantum well to improve the confinement of the carriers, and 15 to 25 nm GaAs capping layers are introduced on both sides of the structure to obtain the right spacer thickness. Silver mirrors were evaporated on both sides of the sample. In order to improve the interface quantity between the silver and the GaAs, a 1.5 nm a layer of Cr metal was evaporated on the GaAs before depositing the silver. The introduction of this layer produces very strong dissipative losses and this reduces considerably the expected inhibition of spontaneous emission. Taking into account these losses, the expected inhibition corresponds to an emission rate reduced only by a factor of 1.6 with respect to the rate in the absence of the cavity. For each of the four samples the photoluminescence decay was

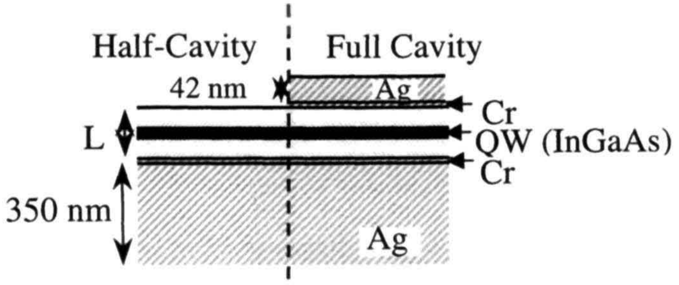

Figure 1: Schematic of microcavity structure. Left-hand side: "half cavity" consisting of semiconductor spacer and a silver mirror on one side only. Right hand side: "full cavity" consisting of semiconductor spacer with a silver mirror on either side. A thin chromium layer is introduced to improve the Ag-GaAs interface.

measured in three different stages of its fabrication process, namely (1) for the as-grown MOCVD spacer on its GaAs wafer, (2) for the spacer transferred on a silver mirror ("half cavity"), and finally (3) for the spacer on a silver mirror plus a 42 nm-thick silver layer evaporated on the free side of the spacer ("full cavity").

The samples were excited optically by a mode-locked Titanium-Sapphire laser tuned to 915 nm, delivering 1.5 ps-long pulses at 4 MHz, with the beam arriving at the sample at an incidence of 70 degrees. The spontaneous emission was collected at normal incidence through a microscope objective and was detected through a time-resolved photon-counting setup. For all samples, photoluminescence decay curves spanning 4 decades of emitted intensity were obtained for a series of 15 different incident intensities corresponding to carrier injection densities between 10^{14} and 10^{18} cm^{-3}. Examples of such decay curves, obtained for the half cavity and the full cavity of sample (b), are given in Fig. 2.

A visual comparison of the luminescence decay curves of the half cavity and the full cavity reveals immediately that the introduction of the top mirror produces a considerable modification in the dynamics of spontaneous emission: the luminescence decay becomes much faster, underscoring the enhancement of spontaneous emission. The decay curves were analyzed in terms of a non-exponential decay that could be fitted to the bimolecular radiative recombination law characteristic of semiconductors

$$\frac{dn}{dt} = -A_{nr}n - Bn_0n - Bn^2$$

where n is the number of carriers injected, A_{nr} is the decay constant for non-radiative recombination, n_0 is the residual doping and B is the bimolecular radiative recombination constant. The value of B extracted from this fit is proportional to the spontaneous emission decay rate obtained from Fermi's Golden Rule. The injected carrier density n was calibrated by determining the incident intensity for which the bimolecular recombination constant B exhibits saturation, when the electron or hole quasi-Fermi levels reach their respective bands.

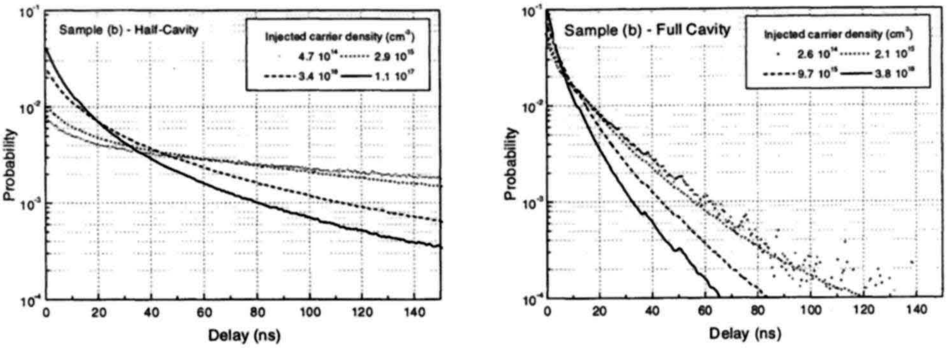

Figure 2: Dynamics of the luminescence decay in sample (b). Left-hand side: spontaneous emission in "half cavity". Right hand side: spontaneous emission in "full cavity".

For all the as-grown samples the B values were of the order of 1.6 x 10^{-10} $s^{-1}cm^{-3}$ as in a quantum well embedded in a bulk semiconductor. For the half cavities the B values ranged between 1.7 x 10^{-10} $s^{-1}cm^{-3}$ and 3.1 x 10^{-10} $s^{-1}cm^{-3}$, corresponding to enhancement by a factor that ranges respectively between 1.1 and 1.9, while for the full cavities the radiative recombination constant ranged from 1.0 x 10^{-10} $s^{-1}cm^{-3}$ to 4.6 x 10^{-10} $s^{-1}cm^{-3}$, corresponding to inhibition by a factor of 1.6 and to enhancement by a factor of 3 respectively. The results are displayed in Fig. 3.

As can be seen in Fig. 3, the measured values for enhancement and inhibition as a function of cavity thickness follow the expected theoretical dependence, calculated by using the "weak coupling" model in which the dissipative losses due to the absorption in the mirrors have been explicitly taken into account by using the experimentally measured reflectivity of the Cr/Ag bi-layers. It should be noted that our results display a systematic deviation whereby the experimental measurements are 25 percent below the theoretical curve. This is probably due to the re-absorption of the emitted light by the quantum well which produces recycling of the excitation and thus changes the apparent emission lifetime, an effect that is not taken into account in our calculations.

The modification of the bimolecular combination constant B by the local electromagnetic environment of the quantum well inside the metallic microcavity in a way similar to that of isolated at terms in the "weak coupling" model is an experimental confirmation that the band-to-band transitions in room temperature semiconductors involve essentially randomly localized dipoles with coherence areas much smaller than the wavelength. At the same time, this result clearly indicates that the bimolecular radiative recombination process is not determined by a kinetic collisional bottleneck, in spite of all the rapid scattering and dephasing processes the carriers undergo at room temperature, but is a true radiative process subject to Cavity Quantum Electrodynamics effects.

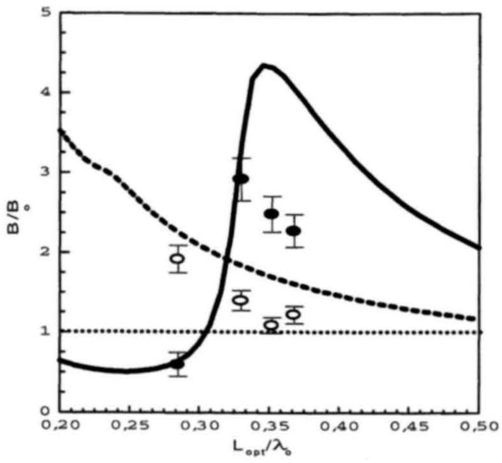

Figure 3: Modification of the bimolecular recombination constant (spontaneous emission rate) of a semiconductor quantum well in a metallic microcavity at room temperature, as a function of the cavity thickness. Full circles: experimental results for full cavities. Open circles: experimental results for half cavities. Full line: theoretical curve for full cavities. Dashed line: theoretical curve for half cavities. Dotted line: spontaneous emission in the absence of mirrors.

6 Conclusion

In conclusion, in our experiments we have demonstrated both enhancement and inhibition of spontaneous emission in room-temperature semiconductor quantum wells placed in microcavities with metallic mirrors. The observation of unambiguous CQED effects, such as enhancement and inhibition of spontaneous emission in room temperature semiconductor microcavities contributes towards the extension of the principles of CQED to complex systems such as semiconductors. This extension is a challenge analogous to that faced by scientists in the early 1960s, when the principles of laser theory, developed initially for isolated atoms or ions, were translated into the language of semiconductors [12] thus opening the way to the development of the semiconductor laser. The observation of CQED effects under conditions similar to those of operating optoelectronic devices (namely, at room temperature and under incoherent injection of the carriers) and the possibility of electrical injection afforded by the metallic mirrors permit us to envisage the design of optoelectronic devices that take advantage of CQED effects, such as high efficiency LED's, ultra-low threshold semiconductor lasers, or emitters producing non-classical light beams with strongly sub-Poissonian photon statistics or controlled trains of photons.

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