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Spontaneous-emission-lifetime alteration in $In_xGa_{1-x}As/GaAs$ vertical-cavity surface-emitting laser structures

Yoshihiko Hanamaki
Research Center for Advanced Science and Technology (RCAST), The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 154, Japan

Hiromu Kinoshita
Department of Electronics Engineering, University of Electro-Communications, 1-5-1 Choufugaoka, Chofu, Tokyo 182, Japan

Hidefumi Akiyama
Institute for Solid State Physics (ISSP), The University of Tokyo, 7-22-1 Roppongi, Minato-ku, Tokyo 106, Japan

Nagaatsu Ogasawara
Department of Electronics Engineering, University of Electro-Communications, 1-5-1 Choufugaoka, Chofu, Tokyo 182, Japan

Yasuhiro Shiraki
Research Center for Advanced Science and Technology (RCAST), The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 154, Japan
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The spontaneous-emission lifetime of $In_{0.2}Ga_{0.8}As/GaAs$ vertical-cavity surface-emitting laser structures was investigated at room temperature as a function of reflectivity of AlAs/GaAs distributed Bragg reflectors (DBR's). As the reflectivity of DBR's became higher, the spontaneous-emission lifetime measured at the resonance wavelength of the cavity became drastically shorter in the cavity axis direction. This spontaneous-emission-lifetime alteration was much more than that of the previous experimental results for quantum wells and the theoretical estimation for atomic dipoles in the planar microcavity structures. [S0163-1829(97)51332-4]

Semiconductor microcavity lasers[1–5] are attracting a lot of attention not only for improving device performances but also for pursuing a fundamental scientific interest, that is, cavity-induced change of the spontaneous-emission lifetime is expected, as originally proposed by Purcell.[6] Vertical-cavity surface-emitting laser (VCSEL) structure which consists of wavelength scale resonators with high reflectivity is a good system for investigating the microcavity effects and for controlling the spontaneous-emission characteristics, since the sample structure with high quality can be systematically varied and monolithically fabricated by molecular-beam epitaxy (MBE) or other advanced crystal-growth methods like metal organic chemical vapor deposition (MOCVD). Yokoyama and his co-workers investigated the spontaneous-emission lifetime of the VCSEL microcavity consisting of three $Al_{0.3}Ga_{0.7}As/GaAs$ quantum wells (QW's) sandwiched by $Al_{0.3}Ga_{0.7}As/AlAs$ and ZnS/SiO_2 distributed Bragg reflectors (DBR's), and reported that the emission lifetime was reduced from 2 ns to 1 ns.[7,8] They also studied microcavities having Rhodamine dye embedded in Langmuir-Blodgett films and reported that a clear spontaneous-emission-lifetime change from 1.8 ns to 1 ns was observed.[9]

Although the spontaneous-emission-lifetime alteration due to the microcavity effect was observed in both cases, the lifetime change was rather small. In addition, theories based on the atomic dipole model have supported such a small change in lifetime in VCSEL structures. Recently, however, Jin and his co-workers have reported an order-of-magnitude enhancement of spontaneous emission of bulk GaAs in a VCSEL structure.[10] They claimed the inadequacy of the standard atomic dipole model, and analyzed the carrier population kinetics in terms of so-called ''beaconing'' and ''Coulomb scattering'' mechanisms.[10]

In this paper, we systematically studied the spontaneous-emission-lifetime alteration of a QW in a series of VCSEL structures at room temperature. We have found significant change of lifetime up to a factor of 6, which is more than the theoretical expectation based on the standard atomic dipole model.

The samples studied were $In_{0.2}Ga_{0.8}As/GaAs$ QW's sandwiched by two AlAs/GaAs DBR's with different reflectivity. In this experiment, the gain width of the $In_{0.2}Ga_{0.8}As/GaAs$ QW at room temperature was designed to be broader than the single cavity mode width, and therefore there was only one cavity mode within the gain width.

Four kinds of VCSEL structures having the same λ-cavity structure and a pair of DBR's with different reflectivity were prepared on (100) undoped GaAs substrates by MBE (VG-80H system). The VCSEL structure grown is schematically shown in Fig. 1. QW's consisting of a 65 Å thick $In_{0.2}Ga_{0.8}As$ well and 1343 Å thick GaAs barrier layers (λ-cavity structure) were fabricated between two sets of DBR's of AlAs/GaAs alternating layers. The layer thicknesses of the AlAs(830 Å)/GaAs(696 Å) DBR were designed to be one-quarter wavelength of the room-temperature emission peak (980 nm) of the $In_{0.2}Ga_{0.8}As/GaAs$ QW. After 3000 Å buffer layers of GaAs were deposited, the bottom DBR was grown at the growth temperature of 580 °C and with V/III ratio of $\sim$8. Then, the λ cavity was grown by keeping the growth temperature at 520 °C to prevent indium desorption and/or

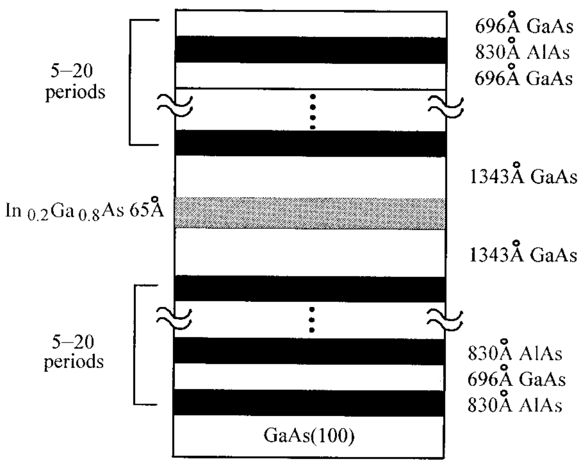

FIG. 1. Schematic diagram of the vertical-cavity surface-emitting laser (VCSEL) structure. The active region ($In_{0.2}Ga_{0.8}As$ single QW) embedded in two GaAs barrier layers is sandwiched between AlAs/GaAs distributed Bragg reflectors (DBR's).

TABLE I. Experimental results and calculated results on maximum reflectivity, stop band width, and linewidth of a slight dip as a function of AlAs/GaAs pair number.

Experimental results (resolution ~8 nm)				
Pair numbers	20	15	10	5
Maximum reflectivity	99.6%	99.0%	98.0%	84.0%
Stop band width (R>98%)	98 nm	97 nm	114 nm	—
Reflectivity at cavity mode	98.6%	94.6%	74.0%	39.3%
Linewidth of a small dip	8 nm	9 nm	10 nm	21 nm
Calculated results (resolution ~8 nm)				
Pair numbers	20	15	10	5
Maximum reflectivity	99.9%	99.9%	99.7%	94.1%
Stop band width (R>98%)	126 nm	127 nm	128 nm	—
Reflectivity at cavity mode	99.2%	84.0%	32.0%	32.0%
Linewidth of a small dip	8 nm	8 nm	8 nm	22.0 nm

segregation. Finally, the top DBR, identical to the bottom mirror, was grown. The pair number of AlAs/GaAs alternating layers was changed from 5 to 20 to investigate the effect of the reflectivity of DBR's on the spontaneous-emission characteristics of the $In_{0.2}Ga_{0.8}As$/GaAs QW. The layers were nominally undoped. The layer thicknesses were calibrated by x-ray diffraction on reference samples.

These samples were characterized by reflectivity, photoluminescence (PL), and time-resolved PL (TRPL) measurements. The reflectivity spectrum measurements were carried out using a quartz-halogen light source and a CdS photoelectric cell in the 800–1200 nm range with the resolution of 8 nm. For the PL spectrum measurements, the excitation light with the wavelength of 860 nm from a clockwise (CW) Ti:sapphire laser was used to excite the $In_{0.2}Ga_{0.8}As$/GaAs QW through the DBR. The PL from $In_{0.2}Ga_{0.8}As$/GaAs QW was detected normal to the sample surface and analyzed by a standard lock-in amplifier system with a Hamamatsu R2658 photomultiplier with the resolution of 0.2 nm. The TRPL measurements were carried out by using a time-correlated single-photon counting system[11] with a Hamamatsu S1 photomultiplier. The VCSEL structures were photoexcited by a mode-locked Styryl 9M dye laser tuned at 860 nm, where the pulse width and the repetition rate were 5 ps and 75.4 MHz, respectively.

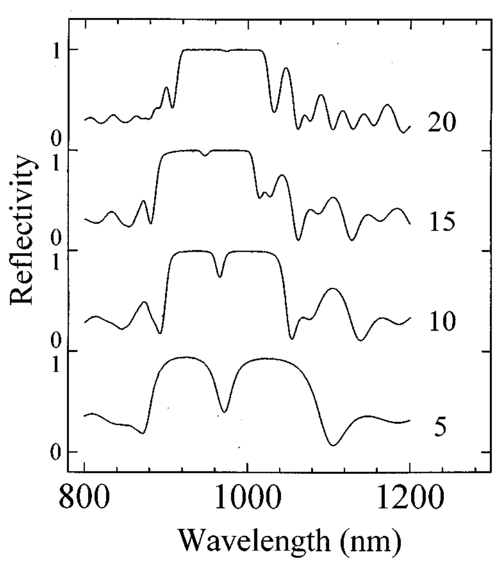

FIG. 2. Reflectivity spectra of each VCSEL structure measured at room temperature.

Figure 2 shows the reflectivity spectra of VCSEL structures with different numbers of AlAs/GaAs pairs measured at room temperature. There are reflectivity plateaus and small dips for all VCSEL structures reflecting characteristics of the Fabry-Perot cavities. It is also seen in this figure that as the number of AlAs/GaAs pairs in DBR's is increased, the maximum reflectivity becomes larger, the stop band width becomes narrower, and the linewidth of the small dip becomes narrower. The results are listed in Table I along with the calculation values. The calculation based on the transfer-matrix method[12] does not include the variation of absorption coefficient over the wavelength rate of interest. The resonance wavelength of the 15-pair sample is seen to be shorter than the others. This is due to the misestimation of the growth rate of GaAs during fabrication and its cavity length became shorter than the designed length (~980 nm).

The PL spectra of the VCSEL structures under the excitation of 80 mW are shown in (1) of Fig. 3. The peak in each PL spectrum coincides with the dip position in the reflectivity spectrum corresponding to the resonance wavelength of the cavity. The cavity length was designed so that the resonance wavelength, 980 nm, becomes equal to the gain peak of the $In_{0.2}Ga_{0.8}As$/GaAs QW. The PL spectrum of the reference $In_{0.2}Ga_{0.8}As$/GaAs QW without DBR formed under the same growth condition which represents the gain spectrum is shown in (2) of Fig. 3. The PL peak of the 15-pair sample is in the shorter wavelength region than the others. However, since the misestimation of the growth rate is only 4%, the gain peak of the 15-pair sample is thought not to be significantly shifted from the wavelength of 980 nm.

It is seen that the PL spectrum shape of the 5-pair VCSEL structure is nearly equal to that of the reference QW. However, as the pair number of DBR is increased, the peak intensity increases,[13] and the linewidth becomes narrower as well, which clearly comes from the microcavity effect. It

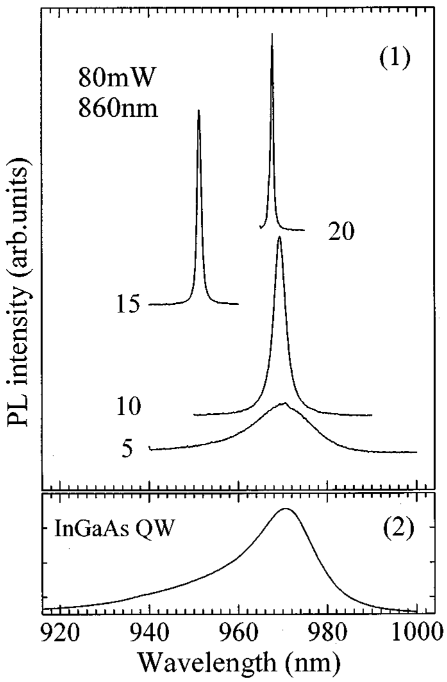

FIG. 3. (1) Photoluminescence spectra for each VCSEL structure measured normal to the epitaxial surface at room temperature. (2) PL spectrum for a single QW without DBR's as a reference.

should be noted that the spectral shape did not change when the excitation intensity was varied from 10 mW to 500 mW, suggesting that the stimulated emission does not participate in the luminescence.

Figure 4 shows the luminescence intensity transient of the samples measured at their resonance wavelength. The decay time of the luminescence intensity of the 5-pair sample is long, 17 ns, which is almost equal to that of the reference QW sample without DBR. The decay time is clearly seen to become shorter with increasing number of AlAs/GaAs DBR pairs. We may attribute this systematic decrease of lifetime

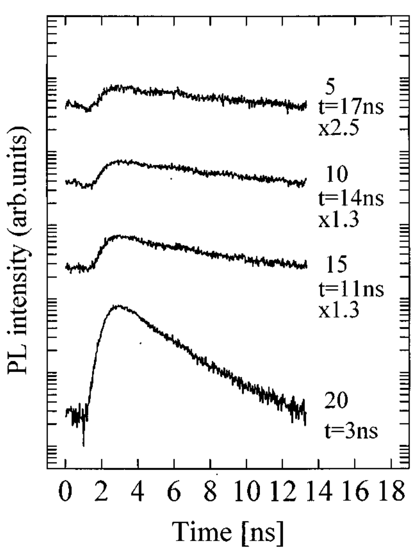

FIG. 4. Luminescence transients of each VCSEL structure measured at room temperature by the time-correlated single-photon counting method.

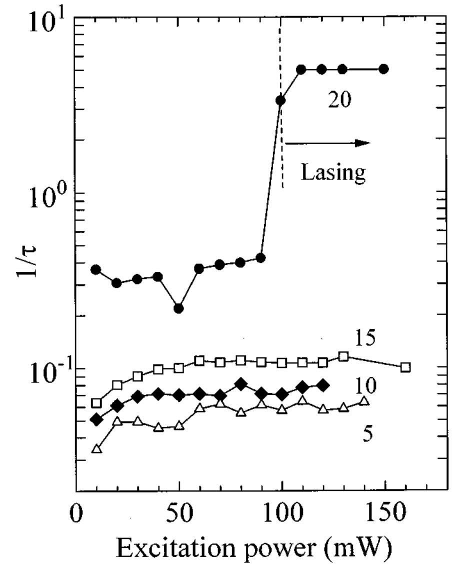

FIG. 5. Excitation power dependence of the inverse of spontaneous-emission lifetime $(1/\tau)$.

to the alteration of the spontaneous lifetime due to the microcavity rather than nonradiative recombination or stimulated emission processes. Note that the lifetime of the 20-pair sample is 6 times shorter than that of the 5-pair sample. Though similar experimental results have been reported for bulk GaAs, such a drastic alteration in the spontaneous-emission lifetime to the best of our knowledge has not been reported for QW's in the microcavity of VCSEL structures.

The excitation power dependence of the emission lifetime was examined, as shown in Fig. 5. It is seen that the inverse of the emission lifetime of VCSEL's with higher reflectivity is always larger under any excitation power. Moreover, the lifetime of each VCSEL does not significantly change between the average power of 40 and 90 mW. This indicates that the stimulated emission effect doe not contribute to the experimental results and that the lifetime shortening comes from the microcavity effect. It is obvious that the abrupt change in the lifetime of the 20-pair VCSEL structure above 100 mW is due to stimulated emission.

To confirm that the drastic alteration in the spontaneous-emission lifetime is due to the high reflectivity of DBR's and hence the enhanced interaction between the excitonic emission and the cavity resonance, we measured the lifetime of another reference sample. It has the same structure as the 20-pair sample, but the cavity length is shortened to have cavity resonance at 930 nm. Since its resonance wavelength (~930 nm) is far from its gain peak (~980 nm), the interaction between excitons and the resonance mode should be small. The emission lifetime of the off-resonant sample measured at the resonance wavelength was found to be 18 ns, comparable to those of samples with low reflectivity or without DBR's, and much longer than that of the on-resonant 20-pair sample. Therefore, it can be concluded that the spontaneous-emission-lifetime change comes from the interaction between the excitonic emission and the cavity resonance.

We comment here on the lifetime (11 ns) of the 15-pair sample which is much larger than that of the 20-pair sample. Since the overlap between the cavity resonance and the gain peak is not perfect, the microcavity effect is not significant compared to the 20-pair sample. If the sample was well fabricated, the lifetime alteration would be more smooth between 10 and 20 of the pair number.

Theoretical explanation for the observed drastic alteration of the spontaneous-emission lifetime of QW's in VCSEL structures has not been achieved yet. However, our present results are very consistent with the recent report[10] by Jin *et al.* It is obvious that the atomic dipole model is not adequate to explain the significant alteration, but that carrier population kinetics in QW should be properly taken into account.

In summary, we fabricated VCSEL structures having the same λ-cavity structure with different reflective DBR's to investigate the reflectivity dependence of the spontaneous-emission lifetime. The PL of these structures showed that the spontaneous emission is enhanced as the reflectivity of DBR's becomes higher. More interestingly, the drastic decrease of the spontaneous-emission lifetime, beyond the previously reported experimental results for QW's and the theoretical estimation based on the atomic dipole model, was observed and attributed to the microcavity effect.

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[13] The integrated intensity of the luminescence probably increased as well with increasing pair number. However, since the real excitation intensity of $In_{0.2}Ga_{0.8}As$/GaAs QW's could not be maintained constant among the samples due to the absorption change of the structure, the integrated intensity increase could not be observed.