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Observation of transient band-gap renormalization in quantum wells

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We have directly measured the carrier-density dependence and the temporal evolution of the band-gap renormalization in a suitably designed $GaAs/Al_xGa_{1-x}As$ quantum-well heterostructure. The transient behavior of the band-gap renormalization is governed by the carrier lifetime. Good quantitative agreement is found with the expected $n^{1/3}$ carrier-density dependence.

Band-gap renormalization (BGR) in semiconductor quantum wells (QW) has been the subject of intense theoretical[1,2] and experimental[3-6] investigations in recent years. These studies made evident the enhancement of the absolute BGR in QW relative to bulk semiconductors and predicted approximately an $n^{1/3}$ carrier areal-density dependence. In addition, a strong well-width dependence of the BGR has been demonstrated[2,3,5] which reduces to an almost universal behavior, provided that the carrier densities are scaled in units of the effective quasi-two-dimensional exciton Bohr radius. The major experimental problem is that BGR is not directly measurable. Information on this important many-body effect can only be obtained by complex multiparameter line-shape fits[3,5] of the luminescence, or by means of delicate pump-and-probe absorption experiments.[4] The spread of the present experimental data is comparatively large,[3,6] which complicates comparison with the most advanced theoretical models. Furthermore, the time dynamics of the transient band-gap renormalization has not been investigated. In this paper, we report direct measurements of the BGR in semiconductor QW. In addition, we provide the first study of the transient band-gap renormalization following the temporal evolution of a nonequilibrium electron-hole plasma. We demonstrate that the strength and the time evolution of the BGR are mainly governed by lifetime effects. Our experimental data agree quantitatively with recent theory.

We investigate two $GaAs/Al_{0.36}Ga_{0.64}As$ QW samples consisting of 25 wells of 10.6 nm width and 15.2-nm-wide barriers. Sample A is grown on a 1-μm-thick $Al_{0.36}$-$Ga_{0.64}As$ layer acting as an optical confiner, while sample B is grown directly onto the absorbing GaAs substrate. The structural parameters of the investigated samples are determined by high-resolution double-crystal x-ray diffraction. We perform time-resolved luminescence experiments under a high photogeneration rate. The light source consists of a frequency-doubled, amplified, actively and passively mode-locked neodymium-doped yttrium aluminum garnet (Nd:YAG) laser providing short pulses of 25 ps full width at half maximum (FWHM) at a 5 Hz repetition rate. The peak power density obtained after focalization over a spot of 800-μm diameter is 0.18 GW/cm^2 The detection system consists of a 25-cm spectrometer, followed by a single-shot streak camera with a two-dimensional readout using a cooled charge-coupled-device (CCD) detector. The overall time resolution is about 20 ps.

The time evolution of the luminescence induced by the intense optical pumping is displayed in Fig. 1 for samples A and B. The different degree of optical confinement results in dramatically different luminescence line shapes.[7] Sample A exhibits a sharp emission line arising in the low-energy tail of the fundamental heavy-hole exciton line (E_{11h}). This band, labeled S, follows the time evolution of the exciting pulse, and decays fast, indicating a very short carrier lifetime ($\tau_A \leq 50$ ps). At longer times, excitonic emission is observed at 799.5 nm (1.553 eV), similar to the low excitation intensity spectra. Sample B exhibits the characteristic high-energy emission due to the progressive filling of the density of states in the QW by the photogenerated electron-hole plasma. The luminescence is spontaneous, and the decay time is $\tau_B \geq 500$ ps. A

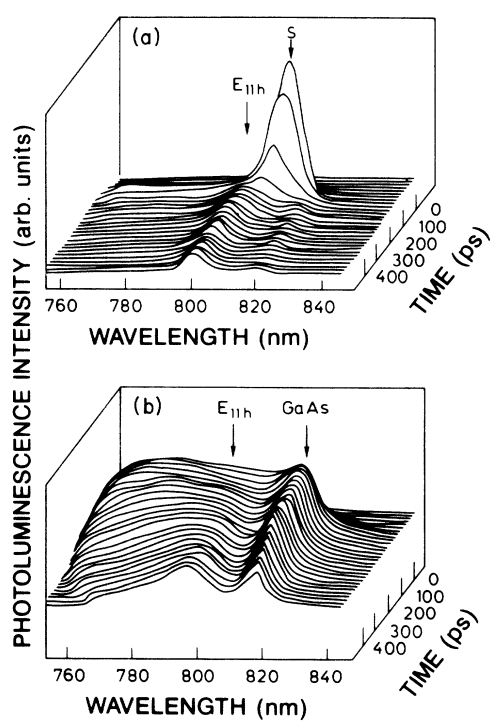

FIG. 1. Time evolution of the electron-hole plasma luminescence in (a) sample A and (b) in sample B at 4 K. The excitation intensity in this case is $I_0/40$. E_{11h} is the $n=1$ heavy-hole exciton luminescence, S is the stimulated emission band, and GaAs indicates the luminescence from the bulk GaAs buffer.

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sharp additional emission is observed around 820 nm (1.515 eV) due to the GaAs buffer. The considerable shortening of the carrier lifetime of sample A is due to the presence of the optically confining $Al_xGa_{1-x}As$ layer, which favors optical amplification of the spontaneous emission at the edge of the renormalized band gap, where self-absorption losses are strongly reduced.[7] No spontaneous emission is observed from higher-energy states in the well, since the short living stimulated emission channel is dominant in all radiative recombination processes.[8,9] The observed luminescence originates from the radiative recombination of the photogenerated electron-hole plasma in both samples. Inelastic excitonic scattering processes cannot be invoked to explain the occurrence of the S band.[10] In fact, in the present experiment the photogeneration rate is 3 orders of magnitude larger than in our previous study[10] (i.e., well above the exciton screening threshold), and spatial homogeneity of the carrier distribution is achieved by using a large excited area on the sample surface.

These results indicate that change in the carrier lifetime causes the different manifestation of the electron-hole plasma luminescence in the two samples, although they consist of identical superlattices. It is then clear that sample A provides a unique tool to selectively investigate the spectral behavior and the temporal evolution of the luminescence emitted at the edge of the renormalized band gap. Hence we can measure the density dependence and the time evolution of BGR in low-dimensional systems by means of luminescence.

The shortening of the carrier lifetime due to the stimulated emission reduces the carrier density in sample A. Neglecting nonlinear absorption processes, the carrier density n is expected to reduce proportional to

$$\frac{\tau I}{\hbar\omega}(e^{-\alpha m L_z}-1),$$

where $\hbar\omega$ is the exciting photon energy (2.41 eV), α the absorption coefficient ($\alpha=1\times10^5$ cm^{-1} at $\hbar\omega=2.41$ eV), m the number of GaAs wells, and the I the excitation intensity. We therefore expect that the observed decrease of the carrier lifetime in sample A reduces the photogenerated carrier density by a factor τ_A/τ_B with respect to sample B under the same excitation conditions. The exact measure of τ_A and its dependence on the excitation intensity are then necessary in order to estimate the actual photogenerated carrier density in the QW. In Fig. 2 we show the typical time decay of the S-band peak. The S emission rises following the time profile of the laser pulse, and the maximum charge density in the well is established with a time constant of about 15 ps. For longer times, the emission intensity follows a single exponential decay $I\simeq e^{-t/\tau_A}$ with a carrier lifetime $\tau_A=31$ ps.

Fitting of the decay curves at different photogeneration rates reveals that the τ_A value is independent of the excitation intensity, provided the intensity threshold for the occurrence of the S band has been overcome. This result allows us to evaluate the effective carrier density in the QW and, in turn to relate the spectral shift of the S band to the photogenerated carrier density, thus getting information on the actual subband renormalization in the well,

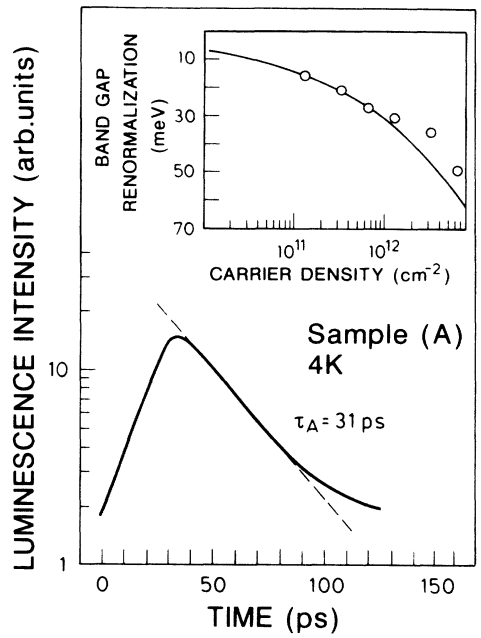

FIG. 2. Time decay of the intensity of the stimulated emission peak of sample A (S band in Fig. 1). The excitation intensity is $I_0/40$, and the temperature 4 K. The broken line is the experimental regression giving a value of $\tau_A=31$ ps. Inset: experimental (points) and theoretical (continuous line) band-gap renormalization vs the carrier density. The experimental BGR values have been taken 70 ps after the end of the exciting laser pulse.

as shown in the inset of Fig. 2. The absolute BGR value has been obtained by comparing the peak energy of the S band with the conduction-to-heavy-hole intersubband transition measured in absorption and in photoluminescence excitation in the same sample (1.562 eV at 4 K). The carrier densities have been estimated by reducing the carrier density obtained from the luminescence line shape of sample B by the factor τ_A/τ_B and linearly scaling with the excitation intensity. The experimental points are compared with the absolute BGR values predicted for a 10-nm GaAs QW by the formula $\Delta E=3.1\times10^{-3}(n)^{1/3}$ meV.[11] The quantitative agreement between theory and experiment is good, despite the simplifying assumption made in the estimate of the absolute carrier density. Large carrier densities lead to a departure from the predicted $n^{1/3}$ dependence, since above 3×10^{12} cm^{-2} the QW gap is renormalized by about 40 meV and merges to the gap of the bulk GaAs buffer. This results in a broad luminescence spectrum which complicates the interpretation of the experimental data.[12] Furthermore, our results are close to the theoretical calculation of Ref. 2 for a 12 nm well width. It is quite evident that an extensive investigation of the well-width dependence of the BGR can easily be performed adopting the above discussed heterostructure configuration, thus allowing a systematic comparison with the theory.

The plot of Fig. 2 differs from the usual luminescence-decay curves, since the energy position of the peaks of the S band changes in time, following the transient behavior

of the electron-hole plasma population in the QW. The effect of the transient carrier density on the BGR is clearly observed in Fig. 3, where we show the fine structure of the spectra of Fig. 1(a) at different photogeneration rates. Low excitation intensity [Fig. 3(a)] generates a broad spontaneous electron-hole plasma emission. The spectrum is characterized by broadened tails caused by the hot-electron carrier distribution on the high-energy side, and by the gap shrinkage on the low-energy side. For longer delays, the electron-hole plasma thermalizes, and the carrier population reduces until excitons are formed after 250 ps. At threshold for stimulated emission, the time evolution of the spectra differs. In the early 100 ps, the sharp S band arises and decays in the low-energy tail of the E_{11h} line. The S emission rising follows the charge-density buildup during the laser pulse. Above the stimulated emission threshold [Fig. 3(c)] the S band rises 25 meV at lower energy than the exciton emission, then blueshifts considerably and disappears in about 150 ps. Two important results should be emphasized: (i) the initial spectral position of the S band depends on the excitation intensity, i.e., on the density of the photogenerated electron-hole plasma. (ii) After the pulse transient the S band blueshifts and smears out until the excitonic luminescence recovers the usual strength.

These data exhibit unique features related to the transient renormalization of the single-particle energy in the photogenerated electron-hole plasma. The spectral position of the stimulated luminescence, which is emitted preferentially at the edge of the gap where self-absorption losses are reduced, closely follows the density dependent shrinkage of the gap. In the early 30 ps, the S band arises at the energy position determined by the initial photogenerated carrier density, i.e., depending on the excitation intensity. For longer times, the net BGR decreases following the time decrease of the carrier density. As a consequence we observe a pronounced blueshift of the luminescence within 150 ps after the exciting pulse.

The temporal evolution of the transient band-gap renormalization is displayed in Fig. 4 for different initial carrier densities. The BGR reaches its maximum value within the first 30 ps, due to the initial maximum photogenerated carrier density. After the extinction of the laser pulse the BGR decreases in time following a $\Delta E \simeq e^{-t/3\tau}$ decay law at all the excitation intensities (dashed curves in Fig. 4). This is consistent with the expected $[n(t)]^{1/3}$ dependence of the BGR,[1,3,13] since the carrier density decays as $n(t) \simeq e^{-t/\tau}$ and the transient BGR must therefore scale as $\Delta E \simeq (e^{-t/\tau})^{1/3}$. The experimental points of Fig. 4 closely follow this exponential decay indicating that the transient subband renormalization takes place in the early 150 ps after the exciting pulse. Inspection of Fig. 4 also

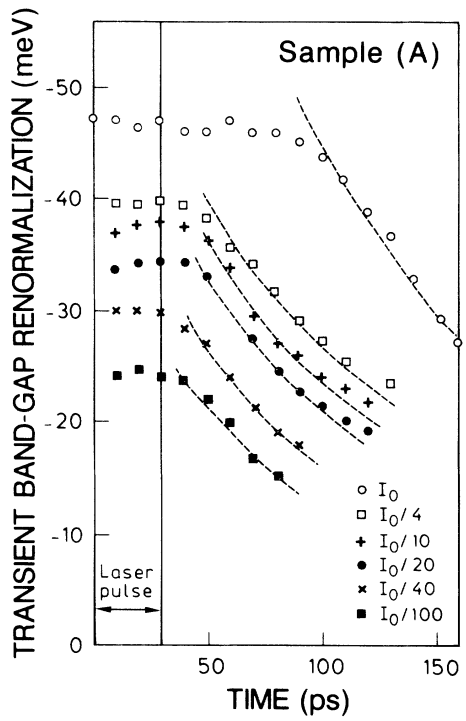

FIG. 4. Transient band-gap renormalization vs time at different photogeneration rates. The vertical line defines the end of the exciting laser pulse. The dashed curves are the exponential regressions with the expected $e^{-t/3\tau}$ decay rate ($\Delta E \simeq n^{1/3}$).

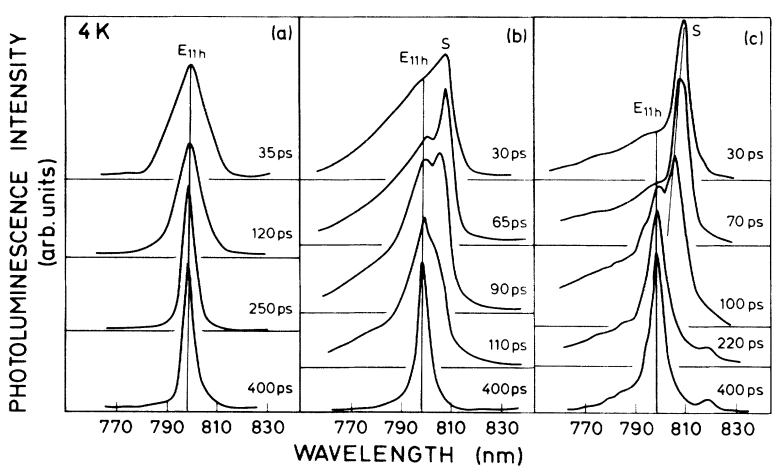

FIG. 3. Time evolution of the S-band peak position at different excitation intensities and at 4 K (a) below the stimulated emission threshold ($I = I_0/400$), (b) at $I_0/100$, and (c) at $I_0/40$. The lines are guides for the eye indicating the E_{11h} exciton and the S-band blueshift.

reveals that at the highest photogeneration rates the transient decay of the BGR is delayed by a few tens of ps depending on the initial carrier density. This is particularly evident in the I_0 curve, where the BGR remains constant up to 60 ps after the exciting pulse extinction. However, we should notice that at such high excitation levels the QW gap has already merged to the bulk GaAs gap, making the quantitative measure of the absolute BGR value difficult. We finally mention that the band-gap reduction measured at different delays Δt well agrees with the expected values, provided the carrier density is scaled as $e^{-\Delta t/\tau}$ (for instance, the experimental points in the inset of Fig. 2 have been taken 70 ps after the exciting pulse). This result is of interest in view of the predicted transient optical bistability due to the increasing absorption bistability at the renormalized band-gap edge.[13]

In conclusion, we have reported the first evidence of transient band-gap renormalization in semiconductor quantum wells. A suitable design of the heterostructure allows us to measure the BGR directly by time-resolved luminescence. We demonstrate that subband renormalization diminishes in time following the time evolution of the photogenerated carrier density. The measured BGR values well agree with current theoretical models either for the absolute BGR value or for the expected carrier density dependence ($\Delta E \simeq n^{1/3}$).

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