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Time-resolved intersubband optical transitions in resonantly optically pumped semiconductor lasers

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Abstract

Using picosecond visible-pump mid-infrared-probe technique, we directly measured the photoexcited electron dynamics in the first conduction subband of a GaAs/AlGaAs quantum well laser, below and above its threshold. Our results clearly show that even at low temperatures, under optical excitation into the lowest heavy hole exciton resonance, GaAs/GaAlAs quantum well lasers lase in accordance with the common two-plasma model and not from an excitonic phase.

Keywords: Picosecond spectroscopy; Electron–hole plasma; Pump and probe; Excitons

The dynamics of charge carriers and photons in operating semiconductor lasing devices has been studied and investigated very intensively during the last few decades [1–4]. It is commonly accepted that in bulk and hetero-structured semiconductor lasers, stimulated emission and gain is achieved by the creation of electron and hole plasmas which the energy separation between their quasi Fermi levels is larger than the semiconductor fundamental band gap [5]. A characteristic signature of this behavior is the spectral shift towards lower energy of the stimulated emission relative to the spontaneous one. In some special cases, however, different behavior was noticed [6]. Under these circumstances, where no such spectral shift was observed, other lasing mechanisms were invoked. Mechanisms such as stimulated emission through annihilation of localized excitons [6–8], exciton–exciton interaction, biexciton decay [9,10] exciton–optical-phonon interaction [11], or even exciton condensation [12,13], have been suggested, and their relevance and agreement with the experimental observations were discussed.

To date, all the experimental studies of these systems were based on optical transitions across the fundamental band gap of the lasing device [1–4,14]. In this work we use, for the first time, optical-picosecond pulses to pump the laser, together with a synchronous infrared (IR) pulse, selectively tuned into the quantum

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structure intersubband optical transition resonance, to directly probe the dynamics of the electronic plasma. In our experiments, the intersubband probe does not create an electron–hole pair but rather induces an optical transition of only one type of photoexcited carrier (an electron in our case). Therefore, the probe pulse is not subjected to the exclusion principle and is extremely sensitive to the momentum state of the carrier whose optical transition is being induced. Thus, a direct measurement of the population of carriers in various momentum states within the first, lasing electronic subband is obtained [15]. In addition, the technique has the advantage of totally different spectral ranges for the pump and the probe beams. This enables a background-free measurements even at resonance, near band-gap pumping.

We study a molecular-beam epitaxy-grown GaAs/AlGaAs heterostructure laser with a periodic quantum structure embedded in its active region. The sample contains 25 periods of 60 Å thick GaAs well followed by 120 Å thick $Al_{0.33}Ga_{0.67}As$ barrier. Two 1 μm thick $Al_{0.5}Ga_{0.5}As$ layers, on each side of the periodic structure separately, confined the optical mode (see inset to Fig. 1). We calculate that at the lasing wavelength (768 nm), 96% of the electromagnetic energy is confined within the active region. Fig. 1 describes schematically the sample. The edges of the sample were cleaved and then polished at 45° to the growth axis in order to form a waveguide for the IR radiation and to enable a considerable electric field component along the growth axis. The substrate was then thinned down and laser bars with 0.6 mm long optical cavities were cleaved. We used a cylindrical lens to focus the pump pulse on the laser bars and monitored the temporally integrated emission along the cavity direction using a monochromator followed by an array of detectors. At the same time, the intersubband photo-induced absorption (PIA) could be determined from the measured transmission of the IR probe pulse through the waveguide. By changing the pump–probe delay time, the temporal evolution of the photoexcited carriers following the excitation pulse was monitored [16]. The temporal and spectral resolution of the experimental setup were 4 ps and 0.8 meV, respectively.

When the focusing lens was oriented perpendicularly to the optical cavity there was no lasing and the integrated emission intensity was almost linear with

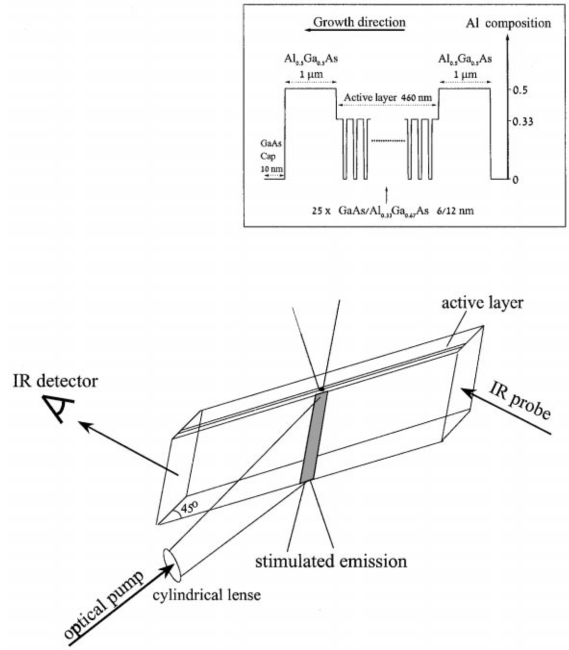

Fig. 1. Schematic description of the experimental arrangement. The pump pulse induces lasing along the cavity direction, while at the same time the intersubband absorption is measured with an IR probe pulse. The inset shows the layers structure.

pump intensity. For a lens orientation along the optical cavity the spectrum evolved with the increase of the excitation density such that above a certain threshold, a low-energy spectral line appeared and lasing action was established. In this case the emission intensity grew vary rapidly with the excitation density above threshold. We estimated a typical threshold density of 4.5×10^{11} cm^{-2} for our laser bars. The lasing emission energy at 10 K was 15 meV below the heavy-hole excitonic resonance (HH1), and 10 meV below the spectral peak of the spontaneous emission.

In Fig. 2 we show the intersubband PIA spectrum (bottom), and its excitation spectrum (top), where the two lowest excitonic resonances HH1, LH1 are clearly resolved. We follow the dynamics of the electronic population in the E1 level, by measuring the PIA transients at various IR wavelengths within the E1–E2 transition resonance. The measured transients are then summed over the whole spectral range of this resonance (8 meV). We pump the sample at the energy of the lowest excitonic

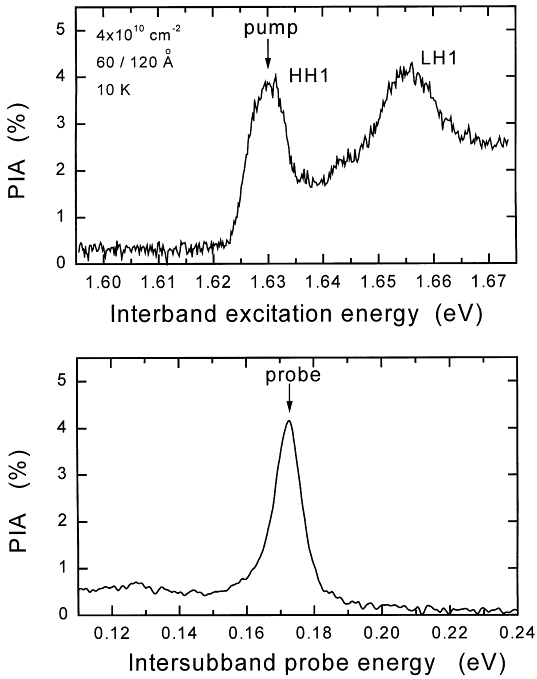

Fig. 2. (bottom) CW measured intersubband PIA spectrum of the laser sample; (top) excitation spectrum of the intersubband PIA measured 60 ps after the excitation pulse.

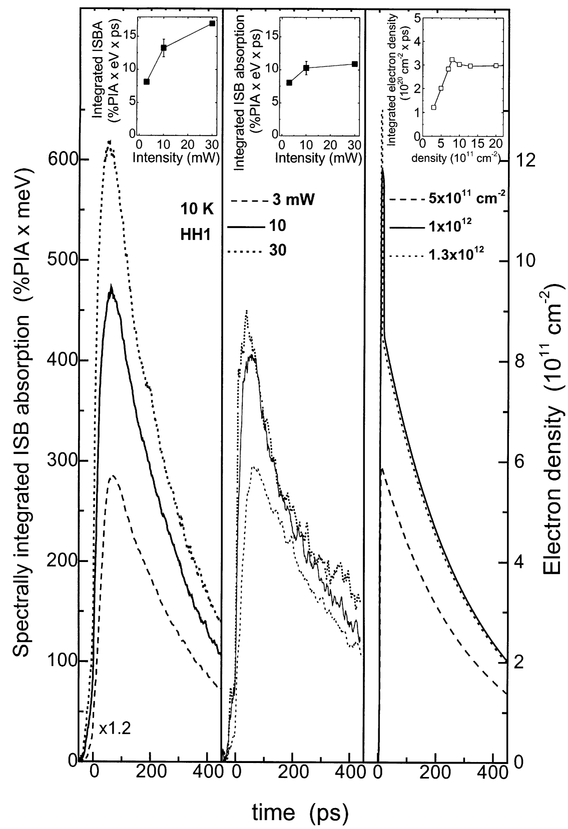

Fig. 3. Spectrally integrated intersubband PIA as a function of time for the "laser" sample at three pump densities: (left (mid) panel) sample in non-lasing (lasing) pumping configuration; (left (mid) inset) temporally and spectrally integrated PIA intensity as a function of pump intensity for non-lasing (lasing) pumping configuration into the HH1 resonance at 10 K; (right panel) calculated electron density from a rate equations model.

resonance, HH1. In Fig. 3 we display the spectrally integrated intersubband absorption as a function of time for three different pump intensities. The left panel shows the temporal evolution under non-lasing conditions. The integrated absorption, which is proportional to the electron density in E1, decays exponentially with a characteristic lifetime of $\cong$ 300 ps, which is almost excitation density independent. The inset (Fig. 3, left panel) displays the temporally and spectrally integrated intersubband absorption as a function of pump density for this case. We note that the average population of E1 grows sub-linearly with pump intensity due to the bleaching of the excitonic resonance at these high excitation densities [17]. The mid panel of Fig. 3 displays the temporal evolution of the PIA when the orientation of the cylindrical lens was along the optical cavity and efficient feedback was therefore set in. Above threshold (10 mW), lasing action was clearly observed. Below threshold (3 mW), the behavior is identical to that of the previous case. At and above threshold, however, the situation is markedly different. In these cases the stimulated emission significantly limits the density of electrons within the lasing cavity and as a result the PIA signal ceases to increase with the excitation density. The maximum of the PIA remains merely the same, even when the excitation density is increased by a factor of 3. After reaching its maximum, the absorption rapidly drops below a certain level, from where it continues to decay in a similar way to the non-lasing cases. The inset to the mid panel of Fig. 3 shows the temporally and spectrally integrated intersubband absorption as a function of pump density for this case. We clearly note that above threshold the electronic density is saturated. This saturation is very similar in nature to the well-known above-threshold clamping of the quasi-Fermi level in steady-state operation of semiconductor lasers [14].

Our results can be interpreted in the following way. Right after the above-threshold pump pulse ($\cong$ 10 ps) lasing action begins and the E1 level is forced to depopulate at very short times due to the presence of immense number of cavity photons during the lasing action. After the electron density drops below the lasing threshold (at $\cong$ 60 ps), the depopulation of E1 is still rapid due to stimulated emission during the intermediate regime. After the electron density drops further ($\cong$ 150 ps), spontaneous emission remains the only decay mechanism of the electronic population and the PIA decays similarly to the non-lasing cases.

In order to quantitatively account for these results, we numerically solved three-coupled rate equations for the electrons, holes and photons within our optical cavity. The temporal behavior of the electronic population for various initial pulse densities are displayed in the right panel of Fig. 3. We note that our simple two-plasma rate equations model explains at least semi-quantitatively our experimental measurements. With our limited temporal resolution we cannot resolve experimentally the initial very rapid spike ($\cong$ 2 ps) that is clearly seen in the calculated electron density above threshold. However, the clamping of the electron density above threshold to its threshold value is very clear both in the experiment and in the calculation (Fig. 3, right panel inset). We find that the calculated pump densities that are best fitted to the measurements are in very good agreement with our estimated experimental pump densities, when the exciton bleaching phenomenon mentioned above is taken into consideration.

These measurements and calculations demonstrate the validity and usefulness of our novel experimental method for studying the dynamics of carriers within an operating quantum-well-based semiconductor laser.

Specifically, our results clearly show that even at low temperatures, under optical excitation into the lowest HH exciton resonance, conventional GaAs/GaAlAs quantum well lasers lase in accordance with the common two plasmas model and not from an excitonic phase.

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