

Two-color picosecond experiments on anti-Stokes photoluminescence in GaAs/AlGaAs asymmetric double quantum wells

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Two-color photoluminescence experiments are performed on the anti-Stokes photoluminescence in GaAs/AlGaAs asymmetric double quantum wells. Direct evidence for forbidden absorption is shown, and its many intriguing aspects, particularly the role of long-lived defects, are revealed. Our experiments shed light on the ongoing controversies between many different models. © 1999 American Institute of Physics. [S0003-6951(99)00749-4]

Recently, anti-Stokes photoluminescence (ASPL) in semiconductor heterojunctions and asymmetric double quantum wells was found and its origin is still being hotly debated. The proposed mechanisms include dipole-dipole interaction,¹⁻³ quantum oscillation,⁴ spatial dephasing,^{5,6} the cold Auger process,⁷⁻⁹ and the two-step absorption process.¹⁰⁻¹⁴ Because experimental bases of each model are mostly indirect, the mechanism of ASPL is controversial and even contradictory.

Many works have used the excitation power dependence of ASPL as the basis for their claims of various mechanisms. The fact that the strength of ASPL increases quadratically in low carrier density, and linearly in high carrier density, was used to support the two-step absorption process.¹⁰ Some samples showed a superlinear power dependence and this was used as strong evidence for the cold Auger process.⁹ But, other samples with similar structures studied by other groups showed sublinear power dependence.¹¹ Much confusion comes from the complexity of the ASPL power dependence. In the viewpoint of the two-step absorption process, the density of the intermediate states participating in the two-step absorption process is very sample dependent. Therefore, the nonlinear power dependence alone cannot be used to exclude the two-step absorption because it can be interpreted that the intermediate states are not saturated. The cold Auger process cannot explain the linear power dependence. In the normal band-to-band Auger process, the transition rate must be strongly power dependent. But, it has been known for more than 10 years that the transition rate of an impurity Auger process does not change in some power ranges.^{12,13} Therefore, the linear power dependence of ASPL excludes the band-to-band cold Auger process but not the impurity Auger process.

Time-resolved studies of ASPL give important insight. The fact that the rise time of ASPL is longer than that of normal photoluminescence (PL) indicates that the source of ASPL is extended in time. Both the cold Auger process and the two-step absorption process can predict this result. Therefore, the problem is not resolved and more clear experimental results are needed.

In this letter, we report the result of a two-color time-

resolved ASPL experiment in GaAs/AlGaAs asymmetric double quantum wells at 10 K, which shows direct evidence of the two-step absorption process in ASPL. In the two-step absorption theory, photoexcited carriers lose energy and populate the intermediate states. The carriers in the intermediate states are excited to the barrier by reabsorbing the photons from the laser or normal PL, and finally, give the ASPL. This theory predicts that the below-band-gap photons can give ASPL if the intermediate states are filled by some method, although they might not give PL by themselves.

Our sample consists of 30 periods of intrinsic GaAs/Al_{0.3}Ga_{0.7}As/GaAs (100 Å/300 Å/50 Å) asymmetric double quantum wells separated by 1000-Å-thick Al_{0.3}Ga_{0.7}As barriers. Figure 1 shows the result of PL and PL excitation experiments. We can see the strong narrow-well ASPL (dotted line) although only the wide well was excited (dotted arrow). In this sample, the narrow-well ASPL efficiency normalized to the wide-well PL is about 3% when we excite the wide-well energy levels above the wide-well excitons. We also detected the strength of the narrow-well ASPL by changing excitation energy continuously (solid line). In this ASPL excitation spectrum, we can see the clear wide-well heavy-hole and light-hole resonance.

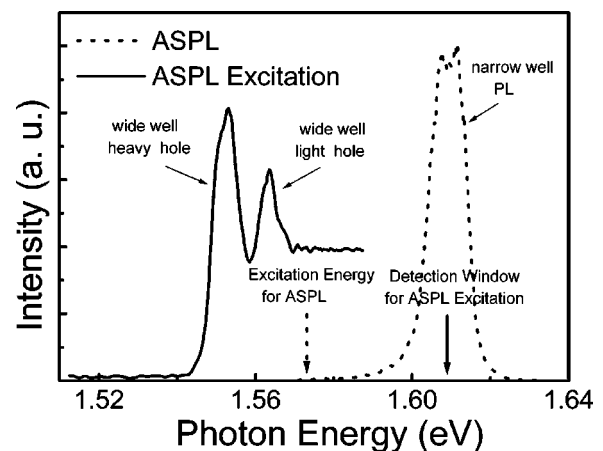


FIG. 1. ASPL (dotted line) from the narrow well when exciting only the wide well (dotted arrow). In this sample, the ASPL/wide-well PL is $\sim 3\%$ when we excite the wide-well energy levels above wide-well excitons. ASPL excitation (solid line) with the detection window at the peak of the narrow-well PL (solid arrow). We can clearly see the wide-well heavy-hole (1.553 eV) and the wide-well light-hole (1.564 eV) resonance.

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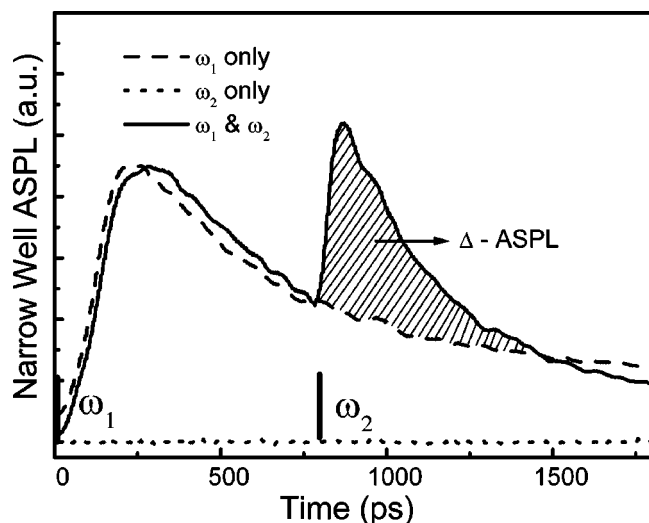


FIG. 2. Time-resolved ASPL of the narrow well when only the ω_1 (1.55 eV, 10 mW) laser, which is resonant with the wide-well heavy-hole exciton is present (dashed lines); when only the ω_2 (1.527 meV, 90 mW), laser, which is below the wide-well band gap is present (dotted lines); and when both beams are present (solid lines).

We used two independently tunable picosecond Ti:sapphire lasers, the first one (ω_1), which was tuned at the wide-well heavy hole to populate the intermediate states in the wide well, and the second one (ω_2), which was tuned below the wide-well band gap to excite only the carriers in the intermediate states. The repetition rate of the lasers is 76.0 MHz and the laser pulse duration is 2–3 ps. The time jitter between pulses from two different lasers was reduced down to ~ 3 ps using the coherent synchroLock system. The narrow-well ASPL was time resolved by streak camera with 10 ps time resolution. We aligned the two laser beams to excite the same spot of the sample using a pinhole and made the ω_2 spot size smaller than that of ω_1 . All experiments were performed at 10 K.

Figure 2 shows the result of the time-resolved ASPL experiment. First, we excited the sample using only the ω_1 tuned at the wide-well heavy-hole energy (dashed lines) and detected the narrow-well ASPL. Second, we excited the sample using only the ω_2 tuned below the wide-well band gap (dotted lines). As expected, we cannot see any narrow-well ASPL in this case. But, the situation dramatically changes when ω_1 and ω_2 coexcite the sample (solid lines). We can see the strong enhancement of the narrow-well ASPL due to ω_2 (hereafter, called Δ -ASPL). We can get the same results when ω_2 is changed down to 1.378 eV. This is direct evidence for the occurrence of forbidden two-step absorption and the resulting ASPL emission in real time. Since Δ -ASPL results from the absorption of the below-band-gap photons, there can be no confusion in terms of distinguishing between the two-step absorption process and all the other mechanisms: for instance, our results are independent of the largely controversial estimates on the cold Auger process cross sections in various samples, because Δ -ASPL is exclusively from the two-step absorption of the below-band-gap photons. We also note that the rise time of Δ -ASPL is much shorter than that of the normal narrow-well ASPL in Fig. 2. In the Δ -ASPL case, the source of ASPL or the ω_2 pulse is localized in time. Therefore, the time evolution of Δ -ASPL is

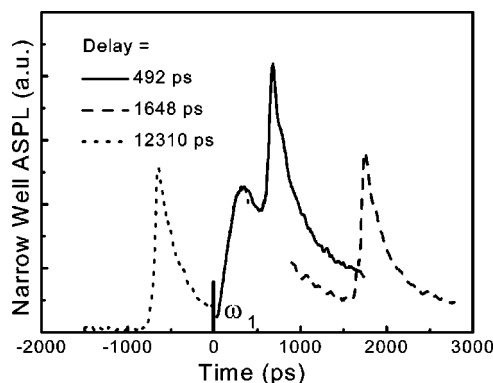


FIG. 3. Time-resolved ASPL of the narrow well at various delays between ω_1 (=1.55 eV, 10 mW) and ω_2 (=1.527 eV, 50 mW). The strength of Δ -ASPL is nearly independent of time delay up to 13 ns.

identical with that of the normal narrow-well PL case.

In Fig. 3, we vary the time delay between ω_1 and ω_2 and probe the change in the Δ -ASPL. Surprisingly, the strength of Δ -ASPL is nearly independent of the time delay up to 13 ns. This excludes the possibility of direct absorption of the below-band-gap photons by excitons, localized or not. This is because in our samples the PL from both free and localized excitons decays with a time constant of 200–500 ps, which is much less than the 13 ns interval within which we can observe substantial two-step absorption. Needless to say, in an ideal, completely defect-free sample, forbidden absorption by excitons, not the trapped carriers, might be important.¹⁴ However, excitons occupy too small a space near the zone center in the momentum space, and therefore, forbidden excitonic absorption might be too small to be experimentally detected. Therefore, we contend that electrons or holes trapped in long-lived microscopic defects make the forbidden transition by absorption of below the band-gap photons. This is because the large momentum of the order of half the Brillouin zone should be provided for the intraband absorption of the photons used in our experiments.

Although the above results confirm that two-step absorption has a strong contribution to ASPL, we cannot conclusively decide whether only the two-step absorption contributes to ASPL or another process coexists. Because the population of the intermediate states is nearly constant during the time from pulse to pulse (~ 13 ns), both the wide-well PL and above the band-gap laser photons (ω_1) can excite the trapped carriers. Considering the longer rise time of ASPL relative to normal narrow-well PL or Δ -ASPL (see Fig. 4), the wide-well PL must be more efficient than laser photons in creating ASPL if only the two-step absorption were to contribute to ASPL. That the wide-well PL is about 100 times more extended in time than the laser pulse can be an explanation if we were to assume that one defect state can be filled and vacated many times over the wide-well PL lifetime. That the Auger process as well as the two-step absorption contribute to ASPL can be another choice of explanation. More study is needed to determine the relative contribution of the two-step absorption process and the cold Auger process, if any.

To summarize, we have demonstrated that the forbidden two-step absorption process contributes strongly to the narrow-well ASPL in our asymmetric double quantum well

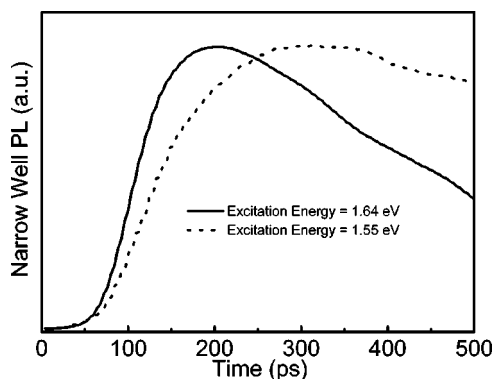


FIG. 4. Time evolution of the narrow-well PL when excitation energy is above the narrow-well band gap (solid line) and the time evolution of the narrow-well ASPL when the excitation energy is below the narrow-well band gap (dotted line).

samples by performing two-color picosecond PL experiments. In addition, we found many aspects of ASPL, such as the important role played by the very long-lived states otherwise invisible. We believe that our work will further stimulate more experimental and theoretical studies on this interesting phenomenon. In particular, exactly what type of defects trap electrons or holes participates in the two-step absorption process is still completely unresolved.

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