

Two-color picosecond and continuous-wave experiments on anti-Stokes and Stokes carrier-transfer phenomena in $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ and $\text{InGaP}_2/\text{Al}_x\text{Ga}_{1-x}\text{As}$ heterostructures

S. C. Hohng, D. W. Khang, Y. H. Ahn, J. Y. Lee, S. Y. Kihm, D. H. Kim, W. S. Kim, J. C. Woo, and D. S. Kim*
Department of Physics and Condensed Matter Research Institute, Seoul National University, Seoul 151-742, Korea

D. S. Citrin

Department of Physics, Washington State University, Pullman, Washington 99164-2814

D. H. Woo, E. K. Kim, and S. H. Kim

Korea Institute of Science and Technology, Cheongryang, Seoul 136-791, Korea

K. S. Lim

Department of Physics, Chungbuk National University, Chungju 360-763, Korea

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We present direct evidence of the two-step absorption process in anti-Stokes photoluminescence in both $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ and $\text{InGaP}_2/\text{Al}_x\text{Ga}_{1-x}\text{As}$ heterostructures using two-color picosecond and continuous-wave photoluminescence experiments. We show information about the lifetime of the defect states that participate in the two-step absorption process. As a result, we conclude that the long-lived states rather than excitons play the dominant role in the two-step absorption process. We also study the possible contribution of the two-step absorption process to Stokes carrier transfer in $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ asymmetric double quantum well structures. [S0163-1829(99)13535-5]

I. INTRODUCTION

Since the discovery of the anomalous Stokes and anti-Stokes carrier-transfer phenomenon in the $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ asymmetric double quantum well structure (ADQW),¹ studies of the carrier-transfer mechanism in semiconductor heterostructure have continued until now, and the generality of these phenomena was made clear. But the interpretations of these phenomena generally do not have a firm ground, and different groups come to contradicting conclusions although they studied similar samples. This situation calls for a different experimental method.

In fact, the anti-Stokes carrier-transfer phenomenon is a general one found in other systems. It has been many years since the anti-Stokes photoluminescence (ASPL) was observed in rare-earth doped ionic crystals,² and in such systems, it is generally accepted that the two-step absorption is responsible for the ASPL. It is still being intensely studied,³ often concerning the energy-loss mechanisms in solid-state lasers and the making of up-conversion lasers.⁴ In indirect band-gap semiconductors such as Si and Ge, ASPL due to the band-to-band Auger process and that involving the transition to deep levels were found.^{5,6}

On the other hand, the agreement on the energy gain mechanism of the anti-Stokes carrier-transfer phenomenon in semiconductor heterostructures has not been achieved.⁷⁻²¹ Though many theories such as dipole-dipole interaction,^{7,8} quantum oscillation,⁹ spatial dephasing,^{10,11} cold Auger process,¹²⁻¹⁴ and the two-step absorption process¹⁵⁻¹⁹ were proposed, the issue is still controversial since these mechanisms are based on such indirect evidences as excitation intensity, pressure, magnetic field, or the temperature dependence of the ASPL.

Most theories resort to the continuous wave (CW) intensity dependence of ASPL. Dipole-dipole interaction, quantum oscillation, and the spatial dephasing models predict linear intensity dependence. The defect-assisted two-step absorption process predicts an initial superlinear dependence followed by a linear dependence.¹⁵ The normal band-to-band cold Auger process predicts nonlinear intensity dependence. Therefore, the authors who support the two-step absorption theory frequently use the linear power dependence as strong evidence against the cold Auger process.^{16,18} However, the Auger process involving deep levels can also have linear power dependence.⁶ Furthermore, since the dynamic range of each experiment differs quite a bit and even the normal PL can have nonlinear intensity dependence, we must be very careful in interpreting the power dependence. Studies on magnetic field and pressure dependence¹⁷ showed an important role of localized states and type-II alignment, but these do not give clear evidence of the two-step absorption process.

Time-resolved studies have shed some important insight into the intricate phenomenon of ASPL.^{15,18,19} In particular, the slow rise time of the ASPL relative to that of normal Stokes PL suggested that the source of the ASPL is temporally extended. However, it should be noted that most of the proposed mechanisms, including the cold Auger process and the two-step absorption process, can result in the long rise time determined by the lifetime of the source.

Another interesting carrier-transfer problem in $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ ADQW is the Stokes carrier-transfer phenomenon. It was found that wide well (WW) PL increases when the excitation laser is tuned at the narrow well (NW) exciton resonances although the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barrier is as thick as 300 Å.¹ This is surprising because the transfer rate deduced from experimental results are orders of magnitude

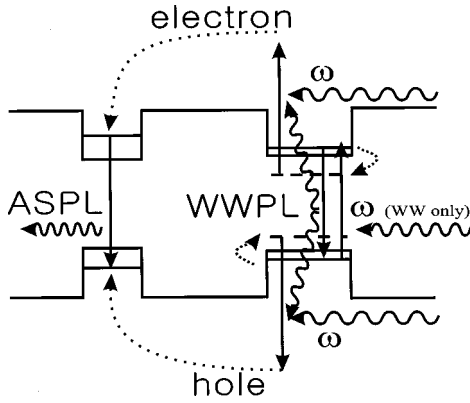


FIG. 1. The mechanism of anti-Stokes PL (ASPL) of asymmetric double quantum wells in two-step absorption theory (WWPL, wide well PL).

larger than the tunneling rate predicted from the one-dimensional tunneling theory and thermal excitation rate over the barrier. Dipole-dipole interaction of excitons,^{7,8} polariton effects,²² reabsorption of the NW luminescence by the WW,²³ and percolation of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barrier²⁴ were proposed as possible explanations for this transfer phenomenon.

As discussed until now, many theoretical models were proposed for the Stokes and anti-Stokes carrier-transfer phenomena, but none of them can give direct experimental evidence of any mechanism, and the issue still remains unresolved.

II. EXPERIMENTAL DETAILS

In this paper, we report the experimental results that enable us to estimate the contribution of the two-step absorption process in Stokes and anti-Stokes carrier-transfer phenomena. In the two-step absorption theory, photoexcited carriers relax to intermediate states and are reexcited over the barrier by absorbing the photons from PL or laser (see Fig. 1). From this picture, we can expect the following phenomena. If the intermediate states are occupied and the energy of photon is large enough to raise the carriers over the barrier, even below the band gap photons can enhance Stokes and anti-Stokes carrier transfer. If we use two lasers, one (ω_1) to populate the intermediate states and the other one (ω_2) with below the band gap energy to reexcite only the carriers in the intermediate states, from the change of PL which is NW ASPL for the anti-Stokes carrier transfer, or WW PL for the Stokes carrier transfer, we can estimate the contribution of the two-step absorption process in Stokes and anti-Stokes carrier-transfer phenomena.

For the time-resolved PL experiment [see Fig. 2(a)], two mode-locked 76-MHz picosecond Ti:Sapphire lasers were synchronized by Coherent SynchroLock system. The PL was time-resolved by Hamamatsu Streak Camera with a time resolution of 10 ps. In all experiments, the spatial overlap of two beams was confirmed by a 100- μm pinhole. We also make the spot size of ω_2 smaller than that of ω_1 to increase the overlap. All experiments were performed at 10 K.

Two different ADQW samples (A,B) and one $\text{InGaP}_2/\text{Al}_x\text{Ga}_{1-x}\text{As}$ were studied for the anti-Stokes carrier transfer. Sample A consisted of 30 periods of intrinsic

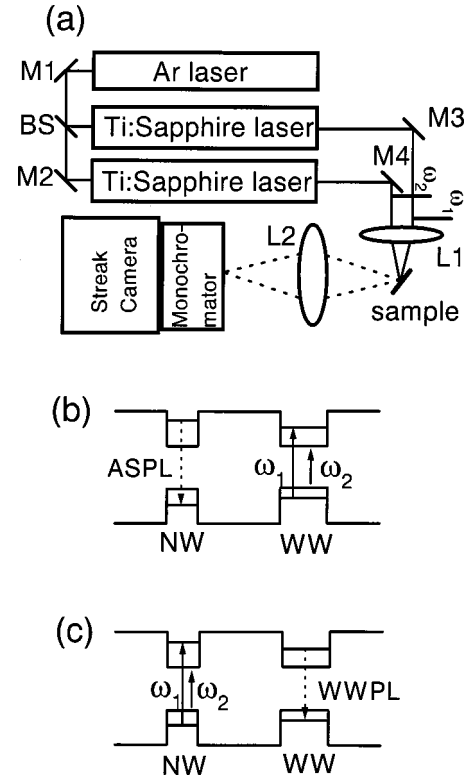


FIG. 2. (a) Schematic diagram of two-color time-resolved PL experiment. (b) Excitation and detection condition for the anti-Stokes carrier-transfer study. (c) Excitation and detection condition for the Stokes carrier-transfer study (NW, narrow well; WW, wide well).

$\text{GaAs}/\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}/\text{GaAs}$ (100 \AA /300 \AA /50 \AA) separated by 1000- \AA thick $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barriers. Sample B had the same structure as that of sample A except that the width of WW was 150 \AA and the inter-well distance was 200 \AA . $\text{InGaP}_2/\text{Al}_x\text{Ga}_{1-x}\text{As}$ sample is the same as the one studied by Cho *et al.*¹⁸ In the Stokes carrier-transfer study, we used many $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ ADQW samples other than sample A and B. Because the experimental results are very similar, we will discuss only sample A.

For the study of anti-Stokes carrier transfer in $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ ADQW, ω_1 is tuned to excite only WW, and ω_2 is tuned below the WW band gap [see Fig. 2(b)]. We studied the change of NW ASPL due to ω_2 at various conditions. For the Stokes carrier-transfer study, ω_1 is tuned at the NW HH energy, and ω_2 is tuned below the WW band gap [see Fig. 2(c)]. We try to probe the change of WW PL due to ω_2 .

For the $\text{InGaP}_2/\text{Al}_x\text{Ga}_{1-x}\text{As}$ heterostructure, the HeNe laser, which has the photon energy between the InGaP_2 band gap and $\text{Al}_x\text{Ga}_{1-x}\text{As}$ band gap, was used as ω_1 . CW Ti:sapphire laser was used as ω_2 , and the change of InGaP_2 ASPL due to ω_2 was probed.

III. RESULTS

A. Anti-Stokes carrier transfer

To characterize the samples, we performed a normal PL experiment. For the $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ ADQW samples, PL excitation (PLE) experiments were also performed. We

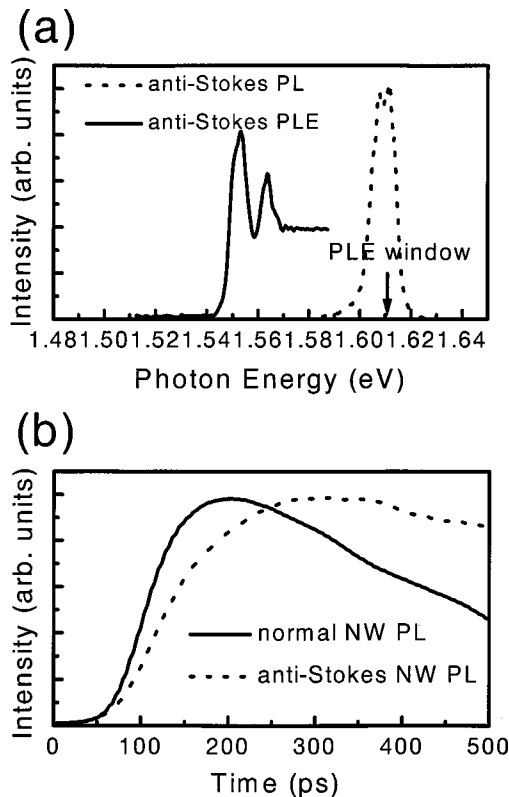


FIG. 3. (a) Anti-Stokes NW PL (dotted line) and anti-Stokes PLE excitation (ASPLE) spectrum with the PL window at the peak of NWPL (solid line) for sample A. The first two ASPLE peaks are the WW heavy-hole (HH) and the WW light-hole (LH) excitons resonance. In this sample, (ASPL/WWPL) is $\sim 3\%$ when we excite the WW continuum. (b) Time evolution of normal NW PL (solid line) and that of anti-Stokes NW PL (dotted line) for sample A.

could detect ASPL from all samples with different efficiency. Figure 3(a) shows the result of sample A. We could estimate that above 3% of carriers excited in WW transfer to NW from the relative strength of ASPL to that of WW PL. Anti-Stokes PLE spectrum shows clear resonances at wide well heavy-hole (WWHH) and wide well light-hole (WWLH) energy.

The time evolution of ASPL is different from that of normal NW PL [see Fig. 3(b)]. ASPL has longer rise time and decays more slowly than normal NW PL. From this result, the quantum oscillation model can be ruled out right away, for it predicts an instantaneous rise. The spatial dephasing model can also be ruled out since it is highly unlikely that any dephasing time could be that long.²⁵ However, as has been stated earlier, this result cannot be used conclusively to pin down the mechanism of the ASPL because the cold Auger process and the two-step absorption process can result in the long rise time determined by the lifetime of the source. But this result has the important implication that gives new light on the ASPL mechanism when combined with results obtained by the two-color picosecond anti-Stokes PL experiment. We will discuss this later.

Figure 4 shows the results of the two-color picosecond PL experiment. For sample A, ω_1 was tuned at WW HH (1.550 eV) and ω_2 was tuned below the WW band gap (1.527 eV). We can see the strong enhancement of ASPL due to ω_2 (hereafter called Δ -ASPL) when ω_1 and ω_2 coexcite the

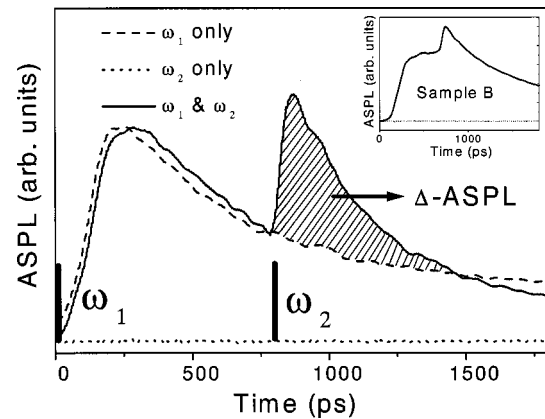


FIG. 4. Time-resolved ASPL of sample A when only the ω_1 (1.55 eV, 10 mW) is present (dashed lines), when only the ω_2 (1.527 meV, 90 mW) is present (dotted lines), and when both beams are present (solid lines). (Inset) The result of the same experiment on sample B grown in a different laboratory. ω_1 is 1.55 eV and 20 mW. ω_2 is 1.512 eV and 60 mW.

sample (solid line) although ω_2 alone cannot make ASPL (dotted line). We obtain the same results when ω_2 was changed down to 1.378 eV. This is the first direct visualization of the two-step absorption process in anti-Stokes carrier-transfer phenomenon. Moreover, generality of the two-step absorption process is shown in the inset of Fig. 4, where the Δ -ASPL also appears in sample B grown in a different laboratory. We also take notice of the difference of time evolution between the normal ASPL and the Δ -ASPL; the rise time of the Δ -ASPL is much faster.

We studied how the time delay between ω_1 and ω_2 affects the strength of Δ -ASPL for sample A (see Fig. 5). Because the strength of Δ -ASPL is proportional to the carrier density in the intermediate states, we can measure the lifetime of intermediate states by measuring the decay of the Δ -ASPL. Surprisingly, Δ -ASPL does not show any hint of decrease up to 13 ns, which indicates that the population of intermediate states is constant in our experimental condition. Thus the possibility of direct absorption of the below the band gap photons (ω_2) by free or localized excitons excited by ω_1 is excluded. This is because in our samples the WW 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.¹⁹ 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 large momentum of the order of half the Brillouin zone should be provided for the intraband absorption of the photons used in our experiments. We should emphasize that this surprising result is very general. Sample B exhibits essentially the same result although it was grown in a different laboratory.

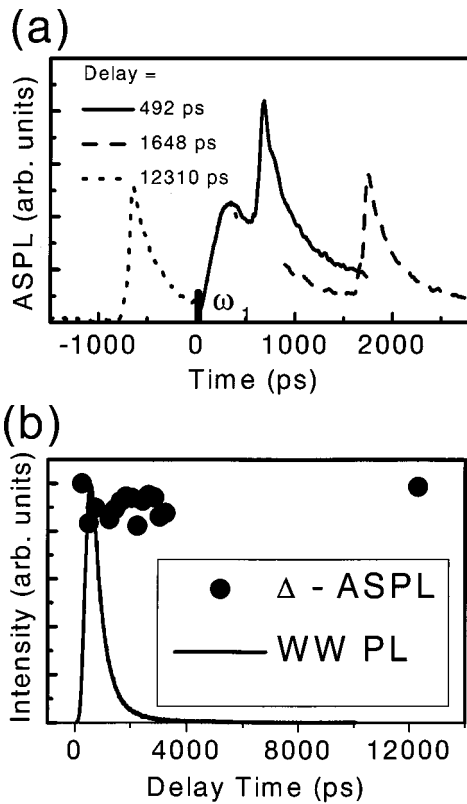


FIG. 5. (a) Time-resolved ASPL at various time delays between ω_1 (1.55 eV, 10 mW) and ω_2 (1.527 eV, 50 mW). (b) The delay dependence of the Δ -ASPL. All data is of sample A. We obtain essentially the same results from sample B.

Until now, the intensity dependence of ASPL has been frequently used for the anti-Stokes carrier-transfer study and simultaneously caused many misunderstandings. We do the same CW experiment on sample A. Sample A showed the transition from the superlinear region to the linear region (see Fig. 6). Similar results have been obtained by many authors who interpreted this as a powerful evidence of the two-step absorption, and the transition point was interpreted as a saturation point of the intermediate states that participate in the two-step absorption process. But this understanding is not as clear as has been thought and much careful treatment is needed.

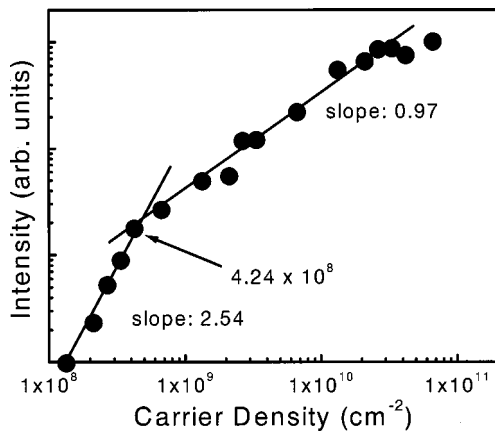


FIG. 6. CW power dependence of ASPL for sample A. We can see the transition from the superlinear region to the linear region.

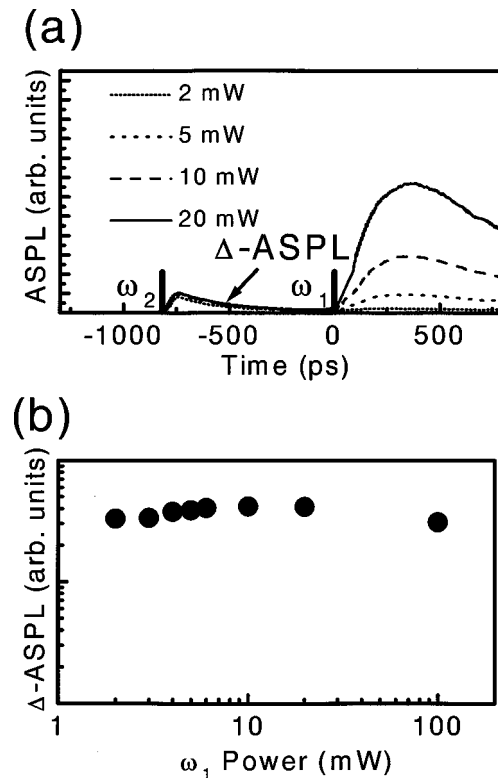


FIG. 7. ω_1 power dependence of Δ -ASPL for sample A. (a) Time-resolved ASPL at various ω_1 power. (b) ω_1 power dependence of the Δ -ASPL. ω_1 photon energy was 1.56 eV. ω_2 photon energy was 1.53 eV, and its power was fixed at 52 mW.

Using the two-color PL experiment, we can study exclusively the two-step absorption process alone in ASPL. The two-color PL experiment has another merit. The power of ω_1 determines only the carrier density in the intermediate states, and the intensity of ω_2 controls the photon flux contributing to Δ -ASPL. Figure 7 shows the result of ω_1 dependence. In this experiment, the time delay between ω_1 and ω_2 was chosen so that ω_2 comes before ω_1 , which means that ω_2 comes ~ 13 ns after ω_1 because the time interval between pulses is about 13 ns. That the strength of Δ -ASPL does not depend on the time delay between ω_1 and ω_2 (Fig. 5) justifies this method. In this configuration, we can see the change of Δ -ASPL more clearly, because Δ -ASPL is separated from the normal ASPL. The strength of the Δ -ASPL does not change when ω_1 power changes from 1 mW to 100 mW. This result indicates that N_c has already reached its saturation value in our experimental conditions at a power as small as 1 mW corresponding to a density of $\sim 10^{10}$ cm^{-2} . Figure 8 shows ω_2 power dependence of Δ -ASPL. We changed ω_2 power with fixed ω_1 power. As expected, Δ -ASPL shows the linear dependence.

Although we found that the two-step absorption process strongly contributes to anti-Stokes carrier transfer from all $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ ADQW samples studied, it is not clear that the two-step absorption process is a general energy gain mechanism that is also found in other systems. The $\text{InGaP}_2/\text{GaAs}$ heterostructure is another frequently studied system for the ASPL. This system is important because of the controversies between the two-step absorption theory and cold Auger process.^{14,16,17} For the experiment, we used the

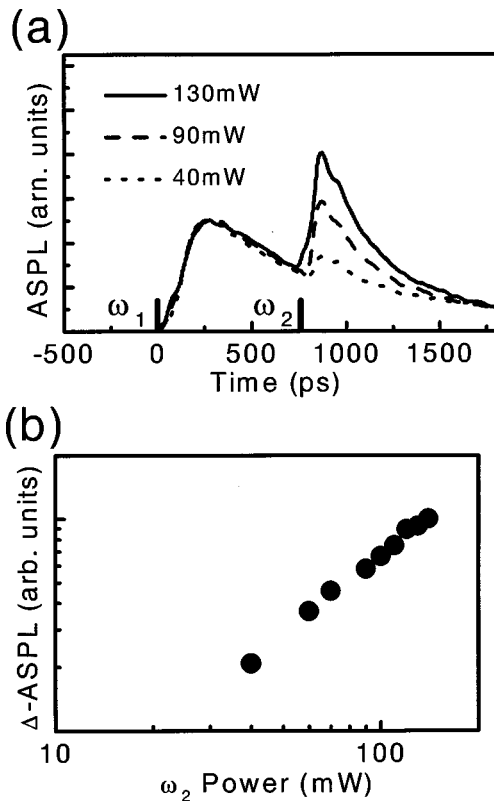


FIG. 8. ω_2 power dependence of Δ -ASPL for sample A. (a) Time-resolved PL at various ω_2 power. (b) ω_2 power dependence of Δ -ASPL. ω_1 photon energy was 1.55 eV, and its power was fixed at 10 mW. ω_2 photon energy was 1.527 eV.

same InGaP₂/Al_xGa_{1-x}As sample that was studied by Cho *et al.*¹⁸ This sample showed strong InGaP₂ ASPL when only the HeNe excites the sample (see Fig. 9). The ASPL is even stronger than the Al_xGa_{1-x}As PL. We probed how the below the band gap photons affect InGaP₂ PL and Al_xGa_{1-x}As PL by coexciting the sample with a CW Ti:Sapphire laser. When Ti:Sapphire coexcites the sample, ASPL was enhanced by $\sim 100\%$. We also noticed that the Al_xGa_{1-x}As PL decreases simultaneously. This feature is generally found in other samples. In the two-color CW PL experiments of GaAs/Al_xGa_{1-x}As ADQW samples, the strength of WW PL

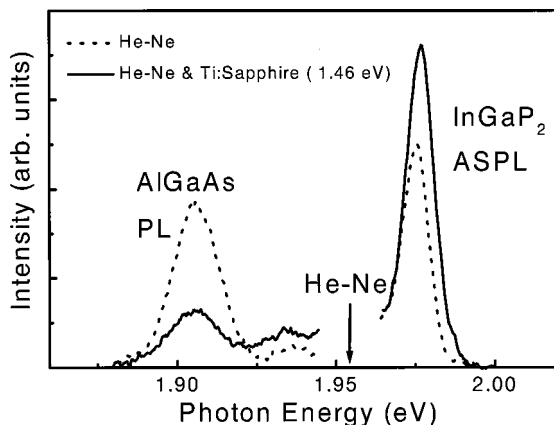


FIG. 9. The result of two-color CW PL experiment in InGaP₂/Al_xGa_{1-x}As heterostructure. ω_1 photon energy was 1.968 eV, and ω_2 photon energy was 1.449 eV.

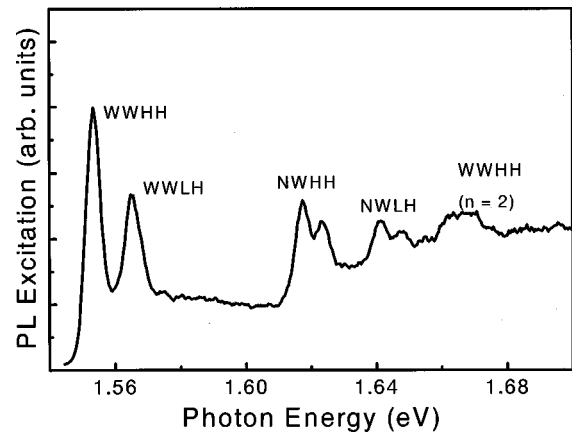


FIG. 10. PL excitation spectrum of sample A with detection window at WW PL. NWHH and NWLH resonance indicates that the carriers excited in NW transfer to WW.

decreased due to ω_2 while the strength of NW PL increased. We believe that electrons and holes are more effectively trapped, and therefore PL intensity decreases because ω_2 evacuates the defect states when ω_2 coexcites the sample.

B. Stokes carrier transfer

PLE spectra have been used to study the Stokes carrier transfer in GaAs/Al_xGa_{1-x}As ADQW.^{1,8,24} Figure 10 shows the results of PLE experiment of sample A. The transfer coefficient per single trial of electron-hole pairs from the NW to the WW can be estimated in the following simple way: when the laser photon energy is tuned at the NWHH, both the NW exciton and the WW continuum are excited. Since the WW luminescence is enhanced by a factor of 2 when the NWHH is resonantly excited, it can be estimated that nearly half of the electron-hole pairs in the WW originate from the NW. We found in absorption experiment that photons are absorbed by NWHH two times more than by WW continuum. From these two facts, we can easily estimate that up to 50% of the resonantly excited NW excitons eventually end up in the WW. With this eventual transfer efficiency T and lifetime of NW excitons (~ 400 ps), we can estimate the transfer coefficient per single trial $t \sim T/N$, where N is the number of round-trips of holes during the NW exciton lifetime.²⁴ Holes were used rather than electrons, since the transfer of holes is generally slower than that of electrons. The transfer coefficient t estimated in this way is of the order of $\sim 10^{-4}$, which is at least 10 orders of magnitude greater than the tunneling coefficient over the one-dimensional mean-field barrier.

To further confirm our interpretation, we performed time-resolved photoluminescence experiments. Data obtained using a streak camera at 10 K are shown in Fig. 11. We first excited WW continuum only and probed WW PL (solid lines). We then excited WW continuum and NWHH simultaneously (dotted lines). The drastic change in time-resolved PL with only a small change of the exciting photon energy again implies the occurrence of a significant transport from the NW to the WW. The fact that WW PL lifetime, which becomes noticeably longer when exciting the WW continuum and NWHH, is nearly the same as that of NW PL

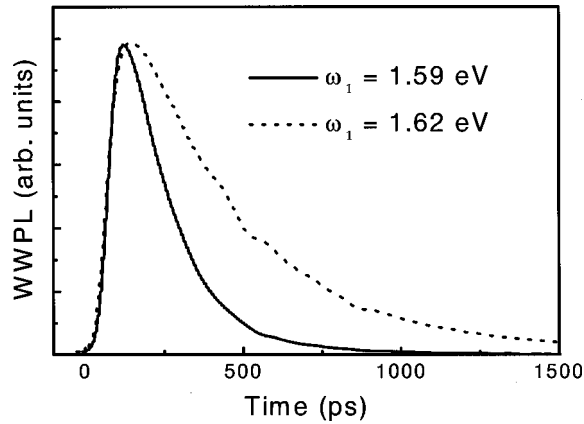


FIG. 11. Time-resolved WW PL at two different excitation energies. The elongated rise time and decay time of WW PL when excited above the NW band gap is a time domain evidence for the carrier transfer from NW to WW.

leaves no doubt that WW PL at later times is dominated by carriers transported from the WW.

We now consider whether the photon-related mechanisms such as the polariton effects, photon reabsorption, or the two-step absorption process contribute, if any, to the Stokes carrier transfer. These mechanisms were proposed theoretically, but there has been no experimental effort to study the possible two-step absorption process or the polariton transfer in the Stokes carrier-transfer phenomenon. We performed the two-color time-resolved PL experiment. As explained before, WW PL was time resolved while ω_1 was tuned at NWHH and ω_2 below the WW band gap. In this experiment, we confirmed the spatial overlap by changing the wavelength of ω_1 after maximizing the strength of Δ -ASPL in the two-color time-resolved anti-Stokes PL setup. Contrary to the case of the anti-Stokes carrier-transfer phenomenon, we could not see any change of WW PL due to ω_2 . It is clear that the contribution of the two-step absorption process in the Stokes carrier-transfer phenomenon is too small to be detected in experiment. Therefore, we can safely rule out photon-related mechanisms. Further studies are needed to unequivocally determine the dominant mechanism for the Stokes carrier-transfer phenomenon.

IV. DISCUSSION

The results of two-color PL experiments on anti-Stokes carrier-transfer phenomenon clearly show the existence of the two-step absorption process. Now, we will discuss whether other mechanisms might also contribute to the ASPL. At first, we will progress on the supposition that only the two-step absorption process contributes to anti-Stokes carrier-transfer phenomenon. We found that the lifetime of intermediate states is so long that the population of intermediate states is nearly constant. Therefore, absorption of both the WW PL and the laser contributes to ASPL in one-color time-resolved PL experiment. We also found that NW ASPL has a longer rise time than normal NW PL. To explain these two facts, we have to conclude that WW PL plays a dominant role in anti-Stokes carrier-transfer phenomenon. To accept this conclusion, a clear explanation is needed as to why

WW PL is more efficient than laser in anti-Stokes carrier-transfer phenomenon.

Intensity difference cannot explain this. Considering the sample thickness and absorption coefficient, we can estimate that about half of laser photons are absorbed by WW. Because the measured quantum efficiency of the sample is a few tens percent, we can safely think that time-integrated laser intensity is larger than that of the WW PL.

WW PL is extended in time although the excitation laser is relatively localized (~ 3 ps). The reabsorption probability of photons is the same whether they are localized in time or not as long as the reabsorption process is not saturated. Therefore, to argue the dominant role of WW PL, the reabsorption process of laser photons must be saturated. In our experimental condition, the reabsorption process is not saturated as shown in Fig. 8.

WW PL has lower energy than excitation laser due to the Stokes shift of the samples. If there is resonance in the two-step absorption process at WW PL energy, the time evolution of NW ASPL should dramatically depend on the excitation energy. When the excitation energy is tuned a few meV below the WW heavy-hole energy, WW PL and laser have the same energy. In this case, laser should be dominant in the anti-Stokes carrier-transfer phenomenon due to the larger laser power than WW PL. Laser is localized in time, and the rise time of ASPL should be short. We did not see the change of rise time but the decrease of ASPL strength.

From the above considerations, it is possible that the two-step absorption is not the dominant process of the anti-Stokes carrier-transfer phenomenon although it is fairly strong. At this time, it is not clear what the dominant process of the anti-Stokes carrier-transfer phenomenon is. But it is clear that the dominant process for the anti-Stokes carrier-transfer phenomenon should be able to explain the following essential features. (1) ASPL shows slower rise time than normal PL. (2) The CW power dependence shows the transition from the super-linear region to the linear region. Although these two facts have been considered as strong evidences of the two-step absorption process, it is not so evident as discussed in previous paragraphs. Therefore, further studies are needed on what the dominant mechanism of the anti-Stokes carrier-transfer phenomenon is.

V. CONCLUSION

To summarize, we have demonstrated the existence of the forbidden two-step absorption process in the anti-Stokes carrier-transfer phenomenon by performing two-color CW and picosecond PL experiments. While the conclusions drawn from one-color experiments have nearly always been controversial, our two-color CW and picosecond experiments leave no doubt as to the existence of the forbidden two-step absorption and subsequent ASPL emission. In addition, we found many interesting aspects of the two-step absorption process, such as the important role played by the very long-lived states otherwise invisible. We believe that our work would further stimulate more experimental and theoretical studies on this interesting phenomenon. In particular,

exactly what type of defects trap electrons or holes and participate in the two-step absorption process is still completely unresolved. Further study on the dominant mechanism of the anti-Stokes carrier-transfer phenomenon is required. As to the Stokes carrier-transfer phenomenon, we clearly showed that photon-related processes such as polariton effect, photon reabsorption, and the two-step absorption process do not play an important role.

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*Electronic address: denny@phya.snu.ac.kr

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