

Perpendicular transport of photoexcited electrons and holes in GaAs/AlAs short-period superlattices: Barrier-thickness and temperature dependence

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Perpendicular transport of photoexcited carriers, which sink into an intentionally enlarged quantum well, is investigated in a set of GaAs/AlAs short-period superlattices with systematically varied AlAs barrier thicknesses as a function of the lattice temperature between 4.2 and 200 K. Excitation-power dependence of the luminescence observed at low temperatures indicates that the ambipolar transport is operative at carrier densities in excess of 10^{15} – 10^{17} cm⁻³. We find that tunneling-assisted hopping conduction prevails at low temperatures via localized states and that the tunneling probability is correlated with the calculated heavy-hole miniband width. We also find a crossover from the hopping conduction to the Bloch-type transport at higher temperatures, which critically depends on the barrier thickness, as a result of thermal activation of carriers to the extended miniband states.

I. INTRODUCTION

Recently, as a result of continuing improvement in the heteroepitaxial-growth techniques, especially in the GaAs-based material, there has been a revival of interest in the field of the carrier transport perpendicular to the quantum-well layers,^{1,2} which was originally discussed by Esaki and Tsu. One of the attractive means to optically study the perpendicular transport in superlattices is the use of an "enlarged well." In short-period superlattices (SPS) with thin barriers, photoexcited carriers sink into an enlarged single quantum well (SQW) which is intentionally introduced.^{3,4} This perpendicular motion of photoexcited carriers from SPS to SQW is utilized to evaluate the conduction along the superlattice direction and has previously been observed by both steady-state and time-resolved photoluminescence experiments.³⁻⁷ The previous experiments, however, were made mostly at the fixed temperatures (4.2 and 77 K) and the barrier-thickness dependence on the tunneling probability in superlattices has not been investigated in detail. The tunneling processes have not been well studied to relate them with the type of carriers (electrons and holes), the lattice temperature, the well and barrier configurations, and the introduced disorder. Recently, Lyo⁸ theoretically investigated temperature dependence of the coherent band diffusion and incoherent well-to-well tunneling processes in an ordered superlattice.

In this paper, the tunneling transport of photoexcited electrons and holes in a series of three GaAs/AlAs SPS samples with systematically varied barrier thicknesses is investigated as a function of the lattice temperature between 4.2 and 200 K. The purpose of this paper is to study the correlation between the temperature-dependent transport processes in superlattices and the miniband width by varying only the barrier thickness. We ob-

served that the SQW luminescence intensity as well as the intensity ratio of SQW to SPS depends strongly on both the barrier thickness and the lattice temperature. On the basis of the detailed luminescence measurements and the Krönig-Penney model calculations of the miniband width, it is inferred that ambipolar tunneling-assisted

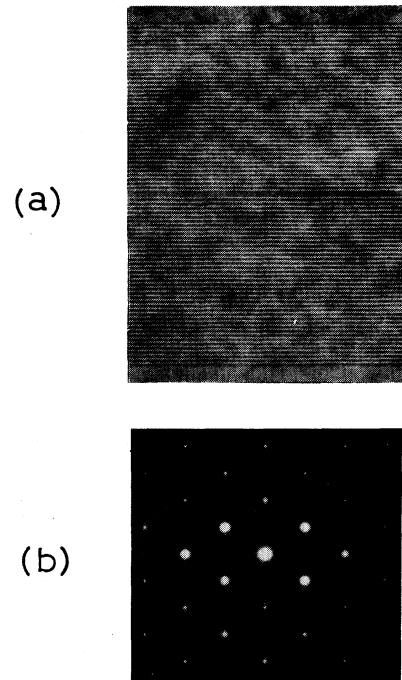


FIG. 1. (a) Cross-sectional image and (b) diffraction pattern by transmission electron microscopy of sample 1, $L_z = 6.36$ nm GaAs SQW confined by GaAs/AlAs SPS ($L_z = 3.18$ nm and $L_B = 0.90$ nm, 40 periods below and above).

hopping conduction prevails at lower temperatures because of the localization caused by thickness fluctuations in the synthesized superlattices. We find that the tunneling probability is correlated with the heavy-hole miniband width which strongly depends on the barrier thickness. With increasing the temperature, we also find a crossover from the hopping conduction to the Bloch-type transport, which critically depends on the barrier thickness, as a result of thermal activation of carriers to the extended miniband states.

II. EXPERIMENTAL

Experiments for the investigation of the perpendicular transport are made with a series of three nominally undoped GaAs/AlAs SPS samples grown on GaAs(100) substrates by molecular-beam epitaxy in a fully computer-controlled Varian Associates Gen II system. Growth conditions optimized in our system for the GaAs/AlAs heterostructures are described elsewhere.⁹ Our nominally undoped thick GaAs samples typically show *p*-type and residual doping levels of low 10^{14} cm^{-3} . The SPS structures consist of 80 periods of the GaAs well with a fixed width (L_z) of 3.18 nm and the AlAs barrier thicknesses (L_B) of 0.90, 1.36, and 1.81 nm for samples 1, 2, and 3, respectively. The thicknesses were determined by small-angle x-ray diffraction experiments. A single GaAs layer in the middle of SPS layers was enlarged to $L_z = 6.36 \text{ nm}$ (SQW) and is served as a sink for photoexcited electrons and holes to monitor the degree of the

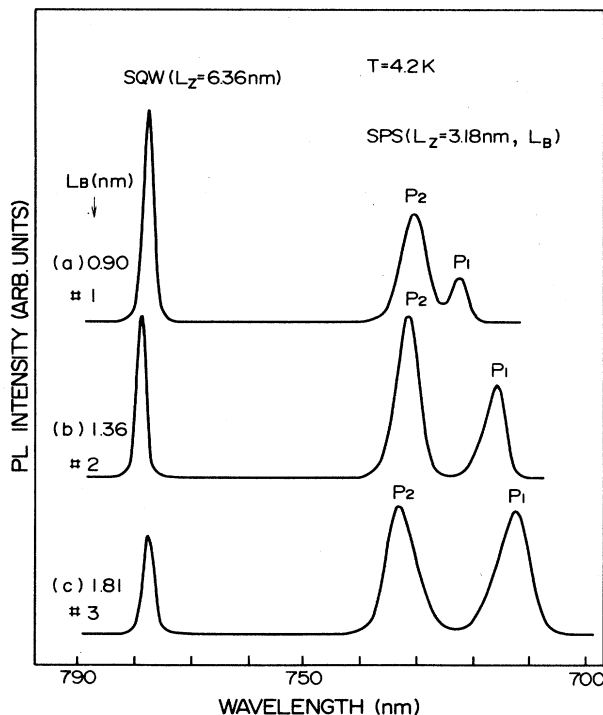


FIG. 2. Photoluminescence spectra of SQW-embedded SPS samples at 4.2 K: (a) sample 1, (b) sample 2, and (c) sample 3. The excitation density is about 10^{14} cm^{-3} .

transport efficiency. The heterostructure was further sandwiched between 0.4 and 0.2 μm $\text{Al}_x\text{Ga}_{1-x}\text{As}$ cladding layers to avoid carrier leakage. As shown in Fig. 1, a cross-sectional image (a) and a diffraction pattern (b) by transmission electron microscopy (TEM) directly confirm designed heterostructures for sample 1, whose barrier thickness is as thin as almost three monolayers. Photoluminescence (PL) measurements were performed in standard lock-in detection systems with a variable temperature cryostat or with a liquid-helium cryostat. For the temperature-dependence measurements, a Kr laser at 647.1 nm was used to uniformly and directly excite the SPS layers since the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layer is almost transparent to the exciting light. The excited carrier density is between 10^{15} and 10^{17} cm^{-3} . A He-Ne laser at 632.8 nm is used for the measurements at 4.2 K for excitation with an estimated carrier density of about 10^{14} cm^{-3} .

III. RESULTS AND DISCUSSION

A. Spectral characteristics and tunneling properties

Figure 2 shows PL spectra for the three SQW-embedded SPS samples at 4.2 K. A sharp PL peak around 780 nm is associated with the $n=1$ electron to

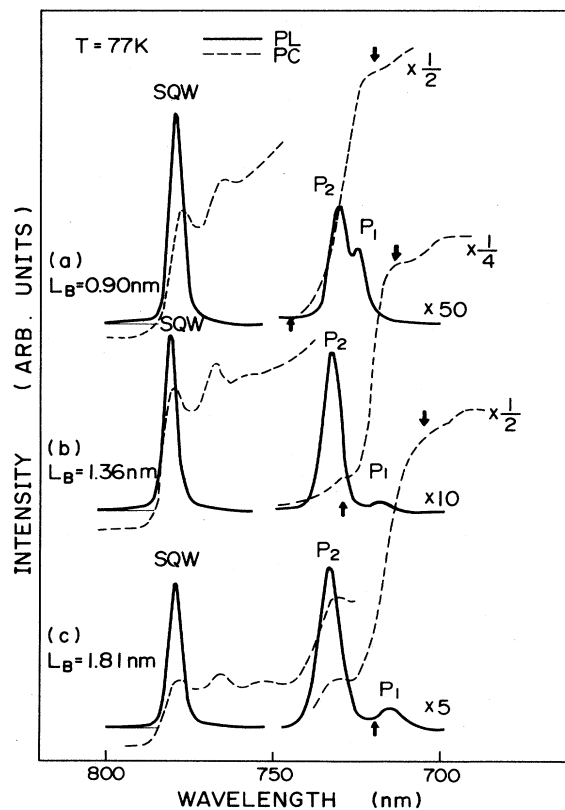


FIG. 3. Photoluminescence (solid curves) and photocurrent (dashed curves) spectra of SQW-embedded SPS samples at 77 K: (a) sample 1, (b) sample 2, and (c) sample 3. The excitation density for the photoluminescence is 10^{17} cm^{-3} .

heavy-hole (1HH) free-excitonic transition of the SQW. Doublet peaks P_1 and P_2 were clearly observed for the SPS structure in the PL spectra of Fig. 2. The splitting energy between P_1 and P_2 peaks was found to be increased from 19 to 49 meV with increasing the AlAs barrier thickness (L_B). The most important variation of emission spectra with L_B in Fig. 2 is the relative increases of the SQW emissions when L_B is decreased. If we note that the volume ratio of SQW to SPS for the excitation is only 2.5%, the transfer of the photoexcited carriers into the SQW is considered to be very efficient and, in fact, strongly depends on the barrier thickness. This result suggests that the transfer is more efficient for thinner barriers as a result of increased overlapping of the quantized wave functions between the neighboring wells. The SQW PL intensity was found to be linearly dependent on the excitation density (10^{15} – 10^{17} cm $^{-3}$) at low temperatures, say 13 K. This implies that both electrons and holes are transferred into the SQW since the photoexcited carrier density ($\geq 10^{15}$ cm $^{-3}$) is larger than the residual hole density ($\sim 10^{14}$ cm $^{-3}$).

Figure 3 shows PL spectra (solid curves) at 77 K for the three SQW-embedded SPS samples, together with in-plane photocurrent (PC) spectra¹⁰ to represent absorption spectral features (dashed curves). A sharp excitonic resonance peak around 780 nm associated with the $n = 1$ electron to heavy-hole free-excitonic transition of the SQW and doublet peaks (P_1 and P_2) for the SPS structure was unambiguously observed in both the PL and PC spectra of Fig. 3. The origin of the doublet peaks was argued in another publication on the basis of polarization properties of luminescence.¹¹ The P_1 peak has mixed heavy- and light-hole characters. The energies of the P_1 peaks correspond to the absorption edge of the SPS and are more strongly dependent on L_B . The absorption edges agree with the calculated miniband edges [indicated by solid vertical arrows (\uparrow) in Fig. 3], based on the Krönig-Penney model calculations assuming 64/36 rules for the band offset. For the calculation we include Bastard's increment of effective-mass discontinuity¹² using the effective-mass parameters from the literature.¹³ The heavy-hole-like P_2 peak is attributed to the localized excitonic transition arising from the layer-thickness fluctuations in the synthesized SPS layers.^{9,11} According to separate experiments¹⁴ to calibrate the 1HH transition energy versus L_z in the GaAs/AlAs system with the barrier thickness of 0.9 nm, the observed P_1 and P_2 splitting

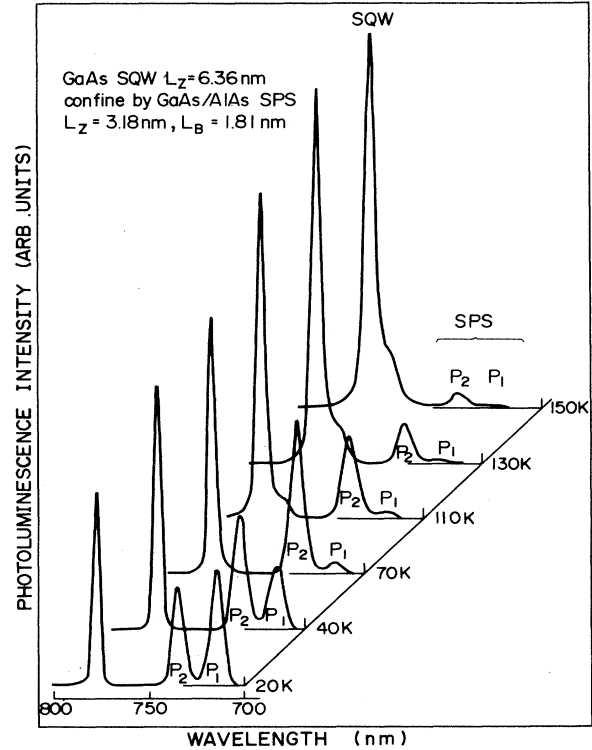


FIG. 4. Temperature dependence of photoluminescence spectra of a SQW-embedded SPS sample (3). The excitation density is 10^{17} cm $^{-3}$.

predicts that the P_2 peak originates from the superlattice states whose well width differs by approximately 1 monolayer. When there exist fluctuations in the layer thickness along the growth direction in the coupled quantum-well system such as in the superlattices, a strong localization of electronic states is theoretically anticipated.^{15,16} Our Krönig-Penney model calculations indicate that fluctuations in the confinement energy by 1-monolayer L_z fluctuations amount to 24 meV, which is in general agreement with the observed splitting of 19 meV for sample 1. The increased P_1 and P_2 separation observed for the thicker barrier samples is also consistent with our assignment of the P_2 peak because of the expected increases of the confinement.

In order to quantitatively estimate the tunneling

TABLE I. Measured values of the intensity ratio $I^{\text{SQW}}/I^{\text{SPS}}$ and calculated values of the miniband widths for electron, light-hole, and heavy-hole in GaAs/AlAs SPS with $L_z = 3.18$ nm and with different L_B parameters. Numbers in parentheses are normalized relative to sample 3.

Sample No.	1	2	3
L_B (nm)	0.90	1.36	1.81
$I^{\text{SQW}}/I^{\text{SPS}}$ (4.2 K)	0.67(4.5)	0.27(1.8)	0.15(1)
$I^{\text{SQW}}/I^{\text{SPS}}$ (13 K)	2.22(5.3)	0.95(2.3)	0.42(1)
$I^{\text{SQW}}/I^{\text{SPS}}$ (77 K)	200(222)	4.6(5.1)	0.9(1)
$2\Delta_e$ (meV)	170(2.5)	107(1.5)	68(1)
$2\Delta_H$ (meV)	18(6.0)	7(2.3)	3(1)
$2\Delta_L$ (meV)	184(2.1)	126(1.4)	88(1)

efficiencies, the intensity ratio of SQW to SPS, $I^{\text{SQW}}/I^{\text{SPS}}$, is evaluated and found to be 0.67 (sample 1), 0.27 (sample 2), and 0.15 (sample 3) at 4.2 K where I^{SQW} and I^{SPS} are the integrated intensity of the SQW and of the SPS (P_1 and P_2) luminescences, respectively. According to the simple rate equation model similar to Chomette *et al.*,⁴ the intensity ratio is related with the transfer time τ_{tr} of the carriers by the equation $I^{\text{SQW}}/I^{\text{SPS}} = f \times \tau^{\text{SQW}}/\tau_{\text{tr}}$, where τ^{SQW} is the radiative lifetime of the SQW and f is a constant factor depending on the radiative and nonradiative lifetimes in the SQW and SPS layers. The transfer is expected to be more efficient for thinner barriers as a result of increased overlapping of the quantized wave functions between the neighboring wells, because the overlapping scaled by the miniband width is proportional to the tunneling probability.² Therefore, the intensity ratio should be proportional to the probability of the tunneling transfer since the transfer time τ_{tr} is inversely proportional to the tunneling probability, assuming that τ^{SQW} does not vary strongly with L_B . To determine relative changes of the tunneling probability by the barrier thickness L_B , miniband widths of electron, heavy hole, and

light hole are calculated based on the Krönig-Penney model. As shown in Table I, it is found that the proportion of $I^{\text{SQW}}/I^{\text{SPS}}$, 0.67:0.27:0.15 (4.5:1.8:1) observed at 4.2 K for samples 1, 2, and 3 corresponds well to the calculated miniband widths of heavy hole, $2 \Delta_H$ (in meV), 18:7:3 (6.0:2.3:1). Parentheses show the proportion relative to the values for sample 3. This result gives additional evidence for the ambipolar transport which is largely governed by the hole transport being consistent with the study of GaAs/Al_xGa_{1-x}As superlattices.¹⁷ As also shown in Table I, the observed ratios at 13 K do not correlate with calculated values of the miniband width for electron and light hole in the SPS and they are also consistent with the results at 4.2 K. The intensity ratio of 0.15 for sample 3 predicts $\tau_{\text{tr}} = 3$ ns when τ^{SQW} is assumed to be 0.4 ns and $f = 1$. This results in a hole velocity of order 3×10^3 cm/s to move the average distance of 100 nm in the SPS layer, which corresponds to the hole mobility of $5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. These results indicate that the perpendicular carrier transport is made possible at 4.2 K by the tunneling-assisted hopping conduction^{8,18} via localized states in the synthesized SPS. At 77 K, however, the SQW PL intensity relative to the SPS emission is found to increase. Thus, the $I^{\text{SQW}}/I^{\text{SPS}}$ ratio increases, and $I^{\text{SQW}}/I^{\text{SPS}}$ values are obtained to be 200 (sample 1), 4.6 (sample 2), and 0.9 (sample 3) at 77 K (see Table I). The increase in the $I^{\text{SQW}}/I^{\text{SPS}}$ value is consistent with the hopping conduction mechanism for samples 2 and 3, although the ratio remains to be an order of unity. However, note that the observed $I^{\text{SQW}}/I^{\text{SPS}}$ ratio for sample 1 increases by almost 2 orders of magnitude. This suggests that a more efficient transport channel is now operative for sample 1 at 77 K than the hopping conduction mechanism. We will discuss temperature dependence of the perpendicular transport in the next section.

B. Temperature dependence

When the lattice temperature was increased, the SQW PL intensity was found to increase significantly. The intensity increases by up to 2.7 times more at 150 K than at 13 K, as is demonstrated in Fig. 4 for the case of the thickest barrier sample (3). This result indicates a substantial increase of the carrier population at the 1HH SQW exciton state, since there is no reason to expect the intensity increase due to an increase of the free-exciton radiative lifetime at higher temperatures.^{19,20} The increase in intensity of the P_2 peak as well as the SQW peak observed in the 13–70 K temperature range in Fig. 4 is attributed to the transfer of carriers from the P_1 emission band. With further increases of the temperature, the SQW intensity still increases at the expense of the P_2 peak. This variation is qualitatively explained by the competition between the carrier transfer time τ_{tr} from the SPS layer to the SQW and the radiative recombination lifetime τ^{SPS} within the P_1 and P_2 emission bands. If the transfer time τ_{tr} is significantly shorter than the radiative lifetime τ^{SPS} , almost all carriers move into the SQW to be consumed by the 1HH SQW free-excitonic emissions before they radiatively recombine in the SPS layer. This is consistent with the experimental observation in

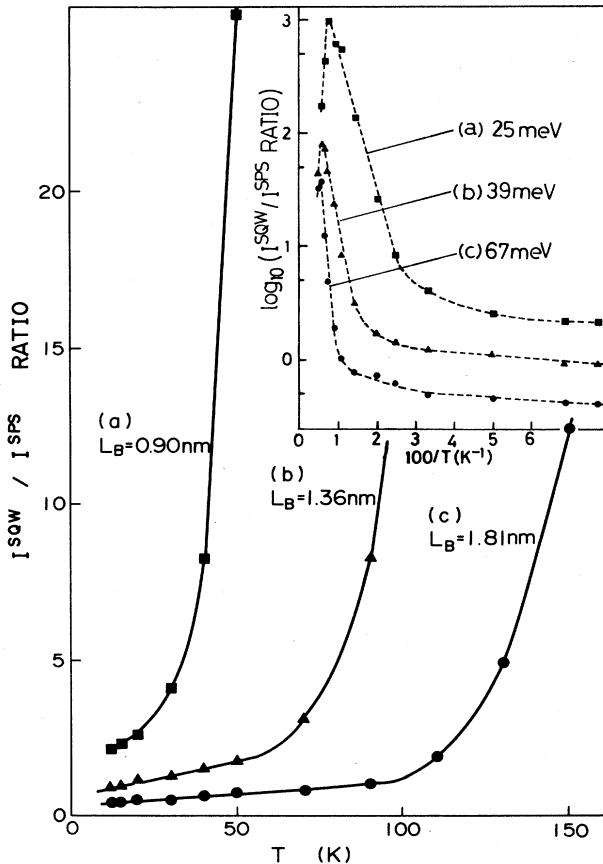


FIG. 5. The ratio of the integrated luminescence intensity of SQW to SPS as a function of the lattice temperature for (a) sample 1, (b) sample 2, and (c) sample 3. The inset shows the semi-logarithmic plot of the ratio against the inverse temperature. Solid and dashed curves are guides to the eye.

Fig. 3 for sample 1 at 77 K. For the case of sample 1, the SQW PL intensity increased only slightly (up to 1.3 times compared to that at 13 K) with the temperature increase because the transfer time is already short enough at 77 K to accommodate the carrier in the SQW.

In order to quantitatively evaluate properties of the temperature-dependent perpendicular transport, the intensity ratio of SQW to SPS, $I^{\text{SQW}}/I^{\text{SPS}}$, is plotted in Fig. 5 as a function of the lattice temperature (T). The results clearly show two different transport processes operative in the temperature ranges studied. At lower temperatures, the luminescence intensity ratios vary with the barrier thickness and they depend linearly on the temperature. These results provide us with further evidence that the tunneling-assisted hopping conduction^{8,18} is predominant at low temperatures via localized states in the synthesized SPS.

With increasing temperature, the ratio $I^{\text{SQW}}/I^{\text{SPS}}$ increases exponentially (see Fig. 5). Thus, photoexcited carriers are very efficiently transferred into the SQW as a result of thermal activation to the extended miniband states. As the barrier thickness was increased, we observed increases of the temperature at which the crossover from the linear dependence on the temperature to the exponential dependence occurred. This can be explained by the increased energy separation of the extended (absorption edge) and the localized (P_2) states as in Fig. 3, since the carriers mostly occupy the localized levels at lower temperatures and require a larger activation energy for the thicker-barrier sample. By plotting the ratio $I^{\text{SQW}}/I^{\text{SPS}}$ as a function of the inverse lattice temperature, a single exponential fit is obtained in the temperature range. The slopes give activation energies, which are estimated to be 25 ± 3 meV, 39 ± 6 meV, and 67 ± 6 meV for samples 1, 2, and 3, respectively. These values are in good agreement with the energy separations, 26 meV (sample 1), 43 meV (sample 2), and 68 meV (sample 3), between the P_2 peak and the absorption plateau (\downarrow) in Fig. 3.

Applying the rate equation model analysis similar to the previous section, the ratio value of 200 measured at 77 K for the thinnest barrier sample (1) predicts $\tau_{\text{tr}} = 10$ ps when the measured value of τ^{SQW} is 1.4 ns (Ref. 21) and $f = 1$. The transfer time τ_{tr} deduced from the rate equation analysis should be taken as the lower limit because the exciton ionization effect on the population at the exciton band is not taken into account. The effect

seems to be more efficient for the case of the SPS layer because the integrated PL intensity for the similar SPS sample without the enlarged well decreases with increasing temperature. If we consider the reduction of the integrated intensity at 77 K, an increase of the τ_{tr} value by a factor of 5 is expected. Consequently, taking $\tau_{\text{tr}} = 10\text{--}50$ ps, we estimate a hole velocity of $10^5\text{--}10^6$ cm/s to move the average distance of 100 nm in the SPS layer. This value is of the same order as the value measured for GaAs/Al_xGa_{1-x}As SPS by Deveaud *et al.*,¹⁷ although the mobility is strongly dependent on the sample. Therefore, such a highly efficient transport observed in the sample with $L_B = 0.90$ nm at 77 K arises from the Bloch-type conduction perpendicular to the heterostructures. For the hopping conduction, on the other hand, the transfer time is reduced by almost 2 orders of magnitude when the $I^{\text{SQW}}/I^{\text{SPS}}$ is comparable to or smaller than unity. The carrier dynamics in the picosecond time domain confirms direct correlation between the carrier flow from SPS and the carrier sink into SQW at 77 K.²¹ A detailed analysis including temperature dependence of such a result will be published in the near future.

IV. CONCLUSION

Perpendicular transport of photoexcited carriers, which sink into an intentionally enlarged quantum well, in a set of GaAs/AlAs short-period superlattices with systematically varied AlAs barrier thicknesses is investigated as a function of the lattice temperature between 4.2 and 200 K. Excitation-power dependence of the luminescence observed at lower temperatures indicates that the ambipolar transport is operative at carrier densities in excess of $10^{15}\text{--}10^{17}$ cm⁻³. We find that tunneling-assisted hopping conduction prevails at low temperatures via localized states and that the tunneling probability is correlated with the calculated heavy-hole miniband width. We also find a crossover from the incoherent tunneling-assisted hopping conduction to the Bloch-type transport at higher temperatures, which crucially depends on the barrier thickness, as a result of thermal activation of carriers to the extended miniband states.

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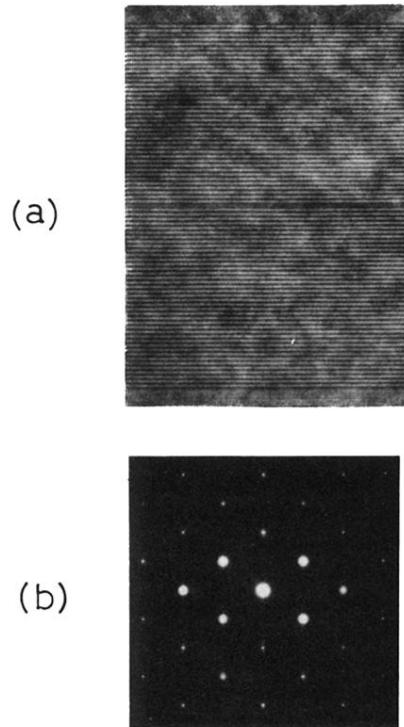


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