Pseudo-negative photocurrent spectroscopy in GaAs-AlAs superlattices

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Pseudo-negative photocurrent spectra, i.e., the appearance of a minimum in photocurrent at an absorption maximum, are experimentally studied at different temperatures and excitation intensities in GaAs-AlAs superlattices on GaAs substrates. Superlattice and substrate are isolated by a thick $Al_{0.3}Ga_{0.7}As$ barrier, but electrically connected through penetrating contacts. A simple model is proposed for the analysis of the conditions which can lead to pseudo-negative photocurrent in this sample configuration. The radiative recombination of the carriers in the superlattice was found to be the main process determining the sign of the photocurrent at an absorption maximum. © *1996 American Institute of Physics.* [S0021-8979(96)09108-7]

I. INTRODUCTION

Photocurrent spectroscopy is often used in order to investigate the absorption properties of semiconductor superlattices (SLs) since it presents fewer technical difficulties than optical absorption measurements. Both parallel[1–10] and perpendicular[11–13] configurations are used. However, the transport properties of the investigated system can turn out to be essential under certain conditions. The counterplay of generation, recombination, and transport properties of the carriers sometimes results in a pseudo-negative or even true negative photocurrent. Pseudo-negative photocurrent (PNPC) refers to the occurrence of a minimum in the photocurrent at an absorption maximum of the SL. However, the photocurrent itself remains positive. Negative photocurrent is defined as the reduction of the current through illumination.

The occurrence of PNPC in perpendicular configuration has been observed in a weakly coupled SL, which is embedded in the intrinsic region of a *p-i-n* diode where both the recombination and the absorption lengths are smaller than the thickness of the intrinsic region. In this sample, only those carriers generated near the electrodes contribute to the photocurrent. Their generation rate, however, can decrease when the absorption length decreases, i.e., when the absorption coefficient increases.[13]

True negative photocurrent in the parallel configuration was observed in InGaAs-InP or InGaAs-GaAs SLs on a GaAs substrate due to a reduction of the mobility of background carriers via Coulomb scattering.[8] In *n*-modulation doped GaAs-AlGaAs quantum well structures, negative photoconductivity can be caused by the interaction with deep donor states.[3] Furthermore, when a sample with an opaque substrate is illuminated and supplied with contacts from the substrate side, the removal of photocarriers from the dominant substrate-related photocurrent by quantum well transitions can also result in PNPC.[9]

In this article, a set of GaAs-AlAs SLs on GaAs substrates with different coupling between adjacent wells is investigated experimentally as well as theoretically in the par-

allel configuration. The SLs are separated from the substrates by very thick barriers, but connected through the alloyed contacts (cf. Fig. 1). Unlike the situation described in Ref. 8, the substrate is opaque at the SL band edge so that the photocurrent of the substrate cannot be neglected. The total photocurrent is always the superposition of the two parallel currents in the SL and the substrate. When the SL is contacted separately from the substrate, the electric field exhibits a perpendicular component since the conducting substrate equalizes the potential between the two contact regions. Hence, a perpendicular current can occur. If this current is large enough, the two subsystems can be connected so that PNPC can even occur under this condition.

A simple model for PNPC in the parallel configuration can be developed in analogy to the perpendicular case.[13] In contrast to the perpendicular case, the total amount of excited electron-hole pairs in the SL as well as in the substrate can contribute to the in-plane photocurrent. The excitation density of carriers decreases exponentially as a function of the distance z from the surface; i.e., it depends only on the direction perpendicular to the motion of the carriers x. We will neglect any possible lateral inhomogeneity of the exciting light within the excitation spot. Since the substrate is considered to be infinitely thick, all photons which are transmitted through the SL are absorbed in the substrate. Therefore, an increase of the number of photons absorbed in the SL leads to a complementary decrease in the substrate. As the transport properties are generally different in both parts of the sample, a change of absorption in the SL may result in a change of the total current. If the recombination length in the SL is, e.g., shorter than in the substrate, the increase of the current in the SL is smaller than the equivalent decrease of current in the substrate resulting in PNPC.[14]

In addition to the photocurrent spectra, the radiative lifetimes have been measured by time-resolved photoluminescence spectroscopy. As far as we can assume a constant electron velocity, these times allow an estimation of the dependence of the recombination lengths on temperature and excitation intensity in order to verify our model. Furthermore, the samples contain a broader single quantum well (SQW) so that perpendicular carrier transfer into this well

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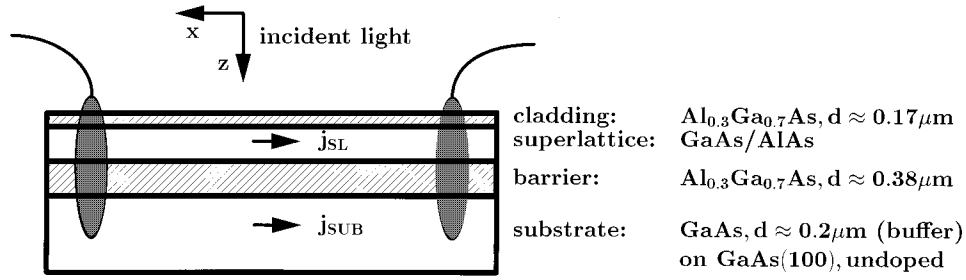
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FIG. 1. Schematic diagram of parallel photocurrent spectroscopy and sample configuration. In most samples the SL contains a single qantum well of different width.

can effect the in-plane photocurrent spectra as a function of the different barrier thicknesses.

The outline of the article is as follows. After a detailed presentation of our model in section II, the experimental results are described (sections III and IV, respectively). In section V the results are discussed in the frame of our simple model.

II. MODEL

The photocurrent is detected in a spectral region where the energy of the excited carriers is always below the barrier height. Hence, the carriers can be assumed to move only in the direction of the external electric field x whereas no current is flowing in the y and z direction. We therefore assume that all quantities such as current, electric field, and electron as well as hole concentrations depend only on x. Although the generation rate can strongly depend on z at the transition energies of the SL only the total generation rate of carriers in the two subsystems (SL and substrate) have to be known. Therefore the empirical model of photoelectric properties, see e.g. Ref. 15, can be written for each subsystem in the following form. It usually contains Poisson's equation

$$\nabla E_{np}(x) = \frac{d}{dx} E_{np}(x) = \frac{e}{\varepsilon}(-n(x) + p(x)), \tag{1}$$

the transport equation (current density j_n due to drift and diffusion, $E = E_{np} + E_{ext}$)

$$j_n(x) = \mu_n e n(x) E(x) + e D_n \frac{dn(x)}{dx} \tag{2}$$

(for electrons), and the continuity equation (stationary state)

$$-\frac{1}{e} \frac{d}{dx} j_n(x) = g - r(x) \tag{3}$$

(for electrons). Similar equations are valid for holes. The recombination rate $r(x)$ is given by

$$r(x) = r_0 n(x) p(x) \tag{4}$$

for radiative recombination, where r_0 is a constant and $n(x)$ and $p(x)$ denote the electron and hole density, respectively. This coupled system (electrons and holes) of nonlinear differential equations can be solved numerically in this simplified form, as well as in the complete form where the quantities depend on all three spatial dimensions.

From the generation rate, which depends on z and is given by

$$g(z) = \alpha(\lambda) g_0 e^{-\alpha(\lambda)z}, \tag{5}$$

the total generation rate of carriers in the two subsystems $g_{SL} = g_0 I_{SL}$ and $g_{SUB} = g_0 I_{SUB}$ with g_0 being a constant can be obtained. The total intensity absorbed in the SL is

$$I_{SL} = I_0[1 - e^{-\alpha_{SL}(\lambda) W_{SL}}] \tag{6}$$

and in the substrate

$$I_{SUB} = I_0 e^{-\alpha_{SL}(\lambda) W_{SL}}, \tag{7}$$

where I_0 denotes the intensity, W_{SL} the total thickness of the SL, and λ the wavelength of the incident light. Note, that both I_{SL} and I_{SUB} depend only on α_{SL} and W_{SL}, since $I_{SL} + I_{SUB} = I_0$.

The system of differential equations is decoupled by the reduction of all recombination properties to the recombination length L so that the continuity equation can be solved separately. The total current is then given by

$$j(\lambda) = j_{SL} + j_{SUB} = g_{SL} L_{SL} + g_{SUB} L_{SUB}. \tag{8}$$

In order to determine whether the photocurrent exhibits a maximum or a minimum at a maximum of the absorption, the second derivative $d^2j/d\lambda^2$ has to be evaluated for $d\alpha/d\lambda = 0$. Using the expressions for $d^2 g_{SL}/d\lambda^2$ and $d^2 g_{SUB}/d\lambda^2$ we obtain

$$\left. \frac{d^2 j}{d\lambda^2} \right|_{\frac{d\alpha}{d\lambda}=0} = g_0 W_{SL} e^{-\alpha_{SL} W_{SL}} (L_{SL} - L_{SUB}) \frac{d^2 \alpha_{SL}}{d\lambda^2}. \tag{9}$$

In this simple picture, the sign of the second derivative of the current at a maximum of the absorption depends only on the difference between the recombination lengths of the SL and the substrate. If the recombination length of the top system, i.e., the SL, is smaller than that of the substrate ($L_{SL} < L_{SUB}$), a maximum in absorption can result in a minimum of the (total) photocurrent. In the case that the recombination length depends on the carrier concentration, PNPC can also occur for $L_{SL} > L_{SUB}$, if the decrease of L_{SL} at an absorption maximum becomes so large that the increase of j_{SL} is smaller than the corresponding decrease of j_{SUB}. For a strong reduction of the mobility, e.g., by Coulomb scattering (cf. Ref. 8), true negative photocurrent can be expected.

Sample	N	L_{GaAs}(nm)	L_{AlAs}(nm)
1	48	5.35	3.0
2	24	5.35	2.0
3	24	5.35	1.0
4	24	5.35	0.5

III. EXPERIMENTS

Photocurrent spectra were measured on GaAs-AlAs SLs embedded by two $Al_{0.3}Ga_{0.7}As$ barrier layers ($d_{top} \approx 0.17$ μm, $d_{bottom} \approx 0.38$ μm) separated from the semi-insulating GaAs substrate by a 0.2 μm buffer layer (cf. Fig. 1). In the following we will not distinguish between the buffer layer and the substrate. The parameters of the SLs are given in Table I. The contacts consist of In alloyed under formation gas atmosphere (20% H_2, 80% N_2) at a temperature of 450 °C. The contact regions are scratched before In is put on in order to guarantee that the contacts of both the SL and the substrate are of sufficiently high quality. Without scratching, even the SL is contacted only partially. Since in this case the quality of the contacts is not reproducible, the total current can vary within one order of magnitude. A measurement on a sample where the contact regions were not scratched, i.e., only part of the SL was contacted, exhibited only weak maxima in the photocurrent (cf. Fig. 2, for the interpretation of the spectra see section IV). After removing the contacts, scratching and alloying the contacts again at the same position, the total photocurrent was about one order of magnitude larger than before, i.e., both the SL and the substrate were now completely contacted. In this case, PNPC was observed for some temperatures [cf. Fig. 3(b)]. This observation confirms the assumption that this effect is strongly connected with the superposition of the parallel currents in the two subsystems.

The photocurrent spectra were recorded for temperatures between 20 K and 120 K with a step width of 20 K under different excitation conditions. The sample was mounted in an optical cryostat (Oxford) where it was illuminated by light
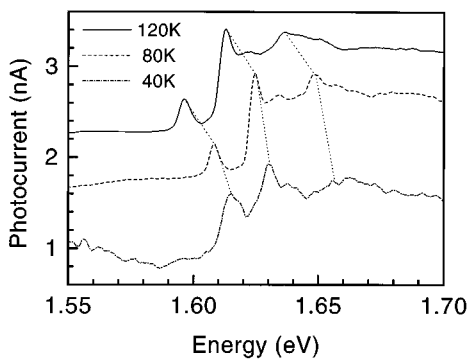

FIG. 2. Photocurrent spectra of sample 3 (only SL was contacted) excited by a Ti:sapphire laser in the nA regime. The spectra at 120 K and 80 K were multiplied by a factor 4 and 2, respectively, and shifted upwards by 2.2 nA and 1.3 nA, respectively, for clarity.
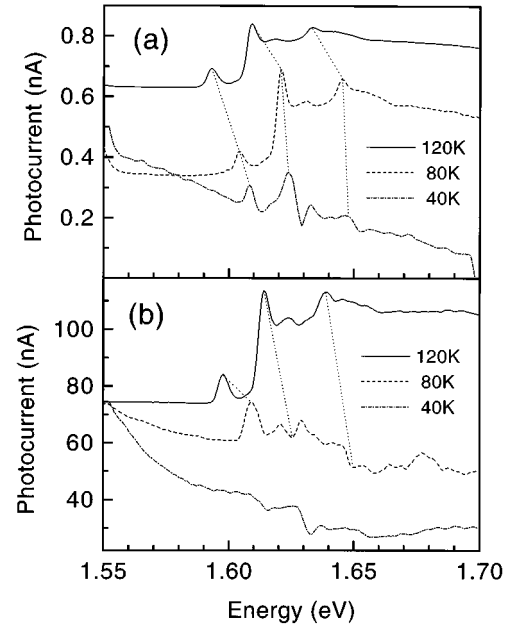

FIG. 3. Photocurrent spectra of sample 3 (SL and substrate were contacted) excited by a halogen lamp in the pA regime (a) and by a Ti:sapphire laser in the nA regime (b). For clarity, the spectra at 120 K were multiplied by 25 (b) and shifted upwards by 0.6 nA (a) and 70 nA (b). The spectrum at 40 K in (a) was multiplied by 0.17 and shifted downwards by 0.6 nA.

from a white light source, i.e., a tungsten halogen lamp with a monochromator (Jobin Yvon), or a tunable Ti:sapphire laser (Coherent). A chopper wheel was able to be placed into the optical axis in order to modulate the light intensity. The circuit consisted of a voltage source (HP), the sample, and a current meter. In the dc mode, when the chopper was removed, the current was measured by a multimeter (HP) whereas in the ac mode a lock-in amplifier (Ithaco) in connection with a pre-amplifier was employed. In the pA region we used the halogen lamp in ac detection mode. The photocurrent spectra were corrected with respect to the lamp/monochromator spectrum. In the nA region the intensity stabilized Ti:sapphire laser was used in ac detection mode as well as in dc detection mode. The excitation intensity of the laser was about two orders of magnitude larger than that of the lamp.

The time-resolved photoluminescence spectroscopy setup consisted of a tunable pulsed Ti:sapphire laser Mira 900 (Coherent) which provided light pulses with a length of 150 fs, an optical cryostat (Oxford), and a streak camera system in combination with a monochromator and a CCD camera as detection system (Hamamatsu).

IV. RESULTS

Figures 2–4 show photocurrent spectra of samples 2 and 3 for three different temperatures (40 K, 80 K, and 120 K) in the ac detection mode. All spectra clearly exhibit the band gap transition (1.63 eV and 1.615 eV at 120 K for samples 2 and 3, respectively) of the SL between the valence band ground state, the first heavy-hole subband (H1), and the first conduction subband (C1) as well as the transition (1.66 eV and 1.64 eV at 120 K for samples 2 and 3, respectively)
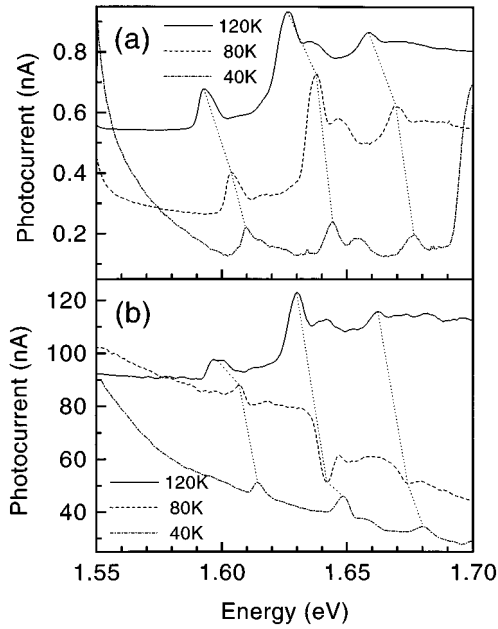
FIG. 4. Photocurrent spectra of sample 2 excited by a halogen lamp in the pA regime (a) and by a Ti:sapphire laser in the nA regime (b). The spectra at 120 K were multiplied by 2 (a) and 15 (b) and shifted upwards by 0.5 nA (a) and 80 nA (b) whereas the spectra at 40 K were multiplied by 0.17 (a) and 0.1 (b) and shifted downwards by 0.7 nA (a).

between the first light-hole subband (L1) and C1. In addition, there is a peak which originates from a broader SQW (1.595 eV at 120 K in the two samples). The H1C1 peak of the SL and of the SQW are also detected in photoluminescence.

The photocurrent spectra of sample 3 do not show any PNPC when the substrate is not well contacted. In this case, the photocurrent in the substrate can be neglected (Fig. 2). If the contacts are extended into the substrate at the same position, this sample clearly exhibits PNPC in the nA regime at 80 K whereas at 40 K the behavior cannot be identified [Fig. 3(b)]. Figure 3(a) shows the photocurrent spectra of this sample in the pA regime. At this low excitation intensity the PNPC disappears. Figure 4 shows the spectra of sample 2 in the pA and the nA regime, respectively. Also this sample exhibits PNPC at 80 K in the nA regime, but only positive photocurrent in the pA regime. The low energy peak of the SQW, however, does not change into a minimum, but remains a maximum. The spectra exhibit a specific behavior in the PNPC regime. Usually, the photocurrent increases with

increasing energy, while in the PNPC regime the photocurrent decreases. However, due to very long time constants of the whole system (sample, pre-amplifier and lock-in amplifier) this unusal behavior can sometimes also occur when positive photocurrent is dominant.

In Table II the results of all samples with regard to the extrema of the photocurrent at the absorption maxima of the SL are summarized. Two tendencies are clearly identifiable. Both decreasing temperature and increasing excitation density lead to PNPC. In order to rule out the effect of different excitation geometries, some photocurrent spectra were additionally recorded with the laser at an excitation intensity comparable to that of the lamp. No significant differences were observed with respect to the lamp spectra.

Figures 5 and 6 show the decay behavior of the photoluminescence for the H1C1 transition of the SL and of the SQW in samples 3 and 2, respectively, at 20 K. In sample 3 carriers are obviously transferred from the SL into the SQW which manifests itself by the initial increase of the photoluminescence intensity of the SQW in the first 500 ps after excitation while the intensity of the SL transition immediately decreases after excitation. This perpendicular carrier transfer into the states of the SQW, which are energetically lower than those of the SL, decreases with increasing barrier thicknesses; i.e., in sample 2 this transfer is much weaker.
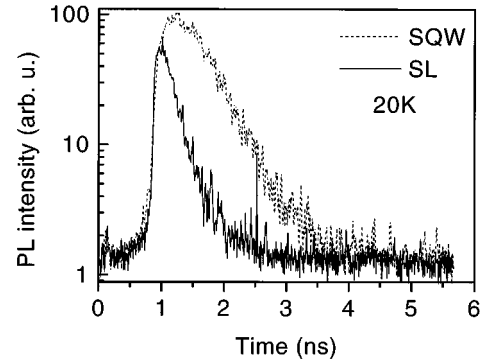

FIG. 5. Radiative decay of the photoluminescence of the superlattice (solid line) and the enlarged single quantum well (dashed line) in sample 3 at 20 K.
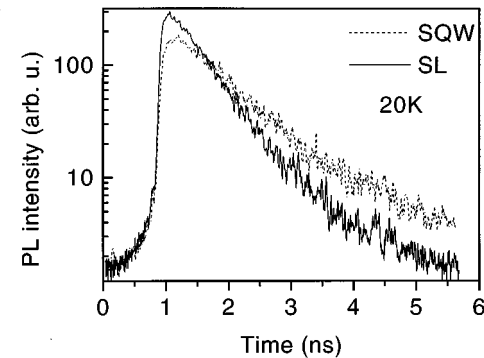

FIG. 6. Radiative decay of the photoluminescence of the superlattice (solid line) and the enlarged single quantum well (dashed line) in sample 2 at 20 K.

TABLE II. Photocurrent extrema at the H1C1 transition, +: maximum (usual behavior), −: minimum (pseudo-negative behavior), 0: not identified.

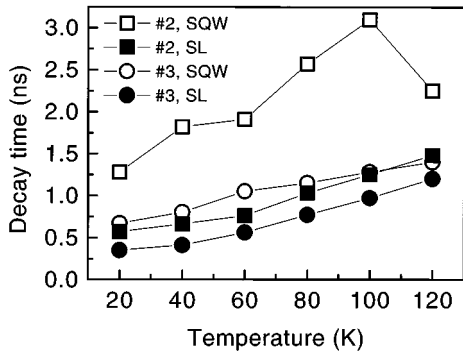| Temp. (K)	1		2		3		4	
	pA regime	nA regime	pA regime	nA regime	pA regime	nA regime	pA regime	nA regime
120	+	+	+	+	+	+	+	+
100	+	+	+	+	+	+	+	0(+)
80	+	−	+	−	+	−	+	0
60	+	0	+	−	0	−	0(−)	−
40	−	0(+)	+	+	+	0	+	−
20	0	0(+)	+	0	+	0	+	−

FIG. 7. Decay times of the photoluminescence of the H1C1 transition of the superlattice and of the enlarged single quantum well in samples 2 and 3 as a function of temperature.

Sample 4 exhibits no transfer due to the absence of such a SQW.

The decay times of the H1C1 transition (SL) as well as of the SQW increase with increasing temperature as shown in Fig. 7. The lifetimes of the SQW are always longer than the ones of the SL. The difference between the lifetimes of the SL and the SQW is larger in sample 2. Furthermore, sample 2 exhibits longer decay times than sample 3. In contrast to SL and SQW, the decay times in the substrate decrease with increasing temperature. In addition, the lifetimes of the SL were found to be longer when the excitation intensity was one order of magnitude lower. For clarity, the data for the substrate and for the lower excitation intensity are omitted in Fig. 7.

V. DISCUSSION

The basis of our model is the superposition of the two photocurrents in the SL and in the substrate. This is clearly shown by the absence of PNPC in sample 3 when the substrate was not contacted. However, contacting the SL separately from the substrate is not a reliable method to avoid PNPC since it was sometimes found even in such samples. In these cases the two currents can be connected by perpendicular components of the electric field due to the deformation of the electrostatic potential. Consequently, the substrate current cannot always be neglected.

In the framework of our model, the positive photocurrent at higher temperatures is equivalent to $L_{SL} > L_{SUB}$ whereas the relation between L_{SL} and L_{SUB} obviously changes at lower temperatures or at higher excitation densities. In the case of PNPC, the recombination length is shorter in the SL than in the substrate. Assuming a constant electron velocity in the two subsystems, the recombination lengths are expected to be proportional to the decay times of the luminescence. In accordance with this assumption, the decay times of the SL were indeed found to decrease with decreasing temperature or increasing excitation density. In the same way, the photoluminescence signal of the substrate decays more slowly at lower temperatures. Furthermore, the decay time of the broader SQW is approximately twice as large as that of the superlattice, which is in agreement with the observation that PNPC has never been found for the SQW. The

main process leading to PNPC in our samples is evidently the decrease of the carrier lifetimes. In contrast to Ref. 8, where Coulomb scattering reduces the mobility of background carriers, the mobility of carriers in our samples even seems to increase with decreasing temperature which is indicated by the fact that at very low temperatures the PNPC vanishes again, although the lifetimes continue to decrease.

The dependence of the recombination lengths on the excitation intensity (larger excitation density is equivalent to shorter recombination time) seems to be a consequence of the fact that the main recombination process is bimolecular recombination of electrons and holes, which depends on the initial intensity. The measurement of the decay times as a function of excitation intensity demonstrates this clear behavior.

The time-resolved photoluminescence measurements suggest perpendicular transfer of electrons from the SL into the SQW. The electrons are trapped there because the energy of the electronic states of the SQW is lower than in the SL. This transfer is only slightly present in sample 2, i.e., the transfer is weaker due to the larger thickness of the barriers. In the frame of our simple model the effect of the perpendicular transport on the PNPC can be understood. The more electrons excited in the SL can move into the SQW, the larger the effective recombination length of the total system, since the recombination time of the SQW is larger than that of the SL. Thus, in the samples which contain an additional SQW (samples 2, 3, and 4) the interpretation of the recombination lengths is more complex. Comparing samples 2 and 3, the PNPC appears at similar conditions (between 60 K and 80 K in the nA regime) although the recombination times are different (cf. Fig. 7). On the one hand, the perpendicular transfer is stronger in sample 3 due to thinner barriers than in sample 2. On the other hand, the recombination times of the SQW are not sufficiently large in sample 3 in order to increase the effective recombination length in the system. These two trends are assumed to compensate each other. In accordance with the shorter recombination time of the SQW in sample 3, its maximum in the photocurrent disappears at 40 K [cf. Fig. 3(b)] while it is always clearly present in sample 2 (cf. Fig. 4). Finally, sample 4 (no perpendicular transfer because of absence of such a SQW) exhibits the PNPC most strongly of all samples.

VI. CONCLUSIONS

In summary, photocurrent spectroscopy in the parallel configuration is a proper method for the investigation of optical properties of semiconductor SLs only, if the SL can be completely isolated from the substrate or it is assured that the recombination length in the SL is large compared to that of the substrate. The latter can possibly be achieved by perpendicular transfer of electrons into a SQW provided that this transfer is sufficiently fast and the recombination length in the well is large enough so that the effective recombination length in the entire system (SL+SQW) is always larger than in the substrate.

ACKNOWLEDGMENTS

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