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OPTICAL NIPI DEVICE CHARACTERIZATION

University of Southern California

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SUMMARY

The motivation of this research project was to develop ultra-sensitive nonlinear optical materials for optical sensor protection, optical signal processing and optical switching. Experiments were performed which measured the response times of quantum well hetero n-i-p-i materials which exhibit optical nonlinearities at intensities of less than a Watt/cm². Also studied were the effects of lateral carrier diffusion in these structures. It was discovered that carriers remain trapped in the quantum wells reducing the magnitude of the transmission change which can be optically induced. Recommendations are made, based on these results, for optimizing device performance. In particular, structures should have very short intrinsic regions and narrow or shallow barriers between the quantum wells.



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PREFACE

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1. INTRODUCTION

Low power nonlinear optical materials are needed for a variety of applications. All-optical spatial light modulators and optical wave mixers (devices which couple energy between two or more optical beams) can be used in information processing, optical interconnects, and sensor protection. For highly parallel operations, intensities of a few milliwatts/cm² per channel are highly desirable, and it is not essential that the response time of the materials be extremely short. By taking advantage of the inherent parallelism of optics, significant information processing can be achieved even if response times are of the order of microseconds. With these requirements in mind, this group has studied hetero n-i-p-i structures which exhibit the desired sensitivity.

For this project, the response times of these materials has been characterized. It was found that material response is in the microsecond to millisecond time regime highly dependent on the illumination and on the initial conditions of the material. To completely describe the decay of photocarriers in a hetero n-i-p-i structure it is necessary to take into account that the photocarriers are distributed throughout the structure, that carriers in different regions have different rates of decay, and that carriers can diffuse laterally out of the photo-spots. In some cases it is possible to simplify this picture considerably. It is shown in Section 2 that a simple model, which treats all the photocarriers in a multiple quantum well hetero n-i-p-i structure as if they had a single lifetime and were concentrated in the doped regions and which ignores lateral diffusion, adequately describes the time response of the structure. In Section 3, excess carrier lifetime is discussed for a CQW H nipi structure in which lateral diffusion can not be neglected. In Section 4, a two lifetime model of excess carrier decay is described which takes into account carrier trapping in the quantum wells of a hetero ni-p-i structure. In the final section, comments are included about the implications of this work for device design.

2. MEASUREMENT OF EXCESS CARRIER LIFETIMES IN A MULTIPLE QUANTUM WELL HETERO N-I-P-I STRUCTURE

The decay rates of carriers photoexcited into the quantum wells of a hetero n-ip-i structure were determined by time-resolved nonlinear transmission measurements. These values are compared with those previously inferred¹ using nonlinear absorption spectra taken under conditions of CW excitation.

The multiple quantum well hetero n-i-p-i (MQW H nipi) sample consists of alternating n-type and p-type Al_{0.3}Ga_{0.7}As layers with eight 100 Å GaAs quantum wells sandwiched between. Details of this structure, pictured in Fig. 1, can be found in Reference 2.



Figure 1. The energy band diagram for the MQW H nipi structure. The built-in electric fields are indicated.

A. Time Resolved Photocarrier Decay - Theory

In this section the results of time resolved measurements of decay are reported. Experimental data are preceded by a discussion of photocarrier lifetime and its relation to measurable quantities.

First a rate equation is derived for the number of photocarriers per unit area in the space charge region of a hetero n-i-p-i sample. The rate at which photocarriers are produced in an intrinsic (absorbing) portion of the hetero n-i-p-i is $I(1-e^{-\alpha L})/hv$ where I is the incident intensity, α is the absorption coefficient of the quantum wells, L is the total length of quantum well material in the i-region, and the units are number per unit area per unit time. It will be assumed here that, before they have had a chance to recombine, all photo-electrons (photo-holes) are swept by the built-in electric fields to the n-type (p-type) material where they compensate space charge. The rate of decay due to recombination we define to be $\sigma_{OW}/\tau_{OW-rec}$ where σ_{OW} is the total number of photocarriers per unit area in the quantum wells of an i-region and τ is identified as a carrier lifetime. The quantity τ_{QW-rec} is not a constant because an electron can recombine with a hole in a different quantum well but the probability of this kind of event depends on the size of the electric field across the i-region. The rate at which photocarriers are swept out of the quantum wells of the MQW H nipi to the doped regions is defined to be σ_{QW}/τ_{out} . The effects of lateral diffusion out of the photo-spots was small for measurements made on this sample.

The net rate of change of σ_{OW} is:

$$\frac{d\sigma_{QW}}{dt} = \frac{I}{h\nu} (1 - e^{-\alpha L}) - \frac{\sigma_{QW}}{\tau_{QW-rec}} - \frac{\sigma_{QW}}{\tau_{out}}$$
(1)

If it is assumed that σ_d , the photocharge per unit area in the doped regions of the n-i-pi, is derived entirely from the flux σ_{QW}/τ_{out} of photocarriers originally created in the quantum wells, then σ_d evolves in time according to:

$$\frac{d\sigma_d}{dt} = \frac{\sigma_{OW}}{\tau_{out}} - \frac{\sigma_d}{\tau_{d-rec}},$$
(2)

where σ_d/τ_{d-rec} is the rate at which excess electrons in the n-region or excess holes in the p-region recombine by thermionic emission over potential barriers or quantum mechanical (unneling through the barriers.

If τ_{out} is very small (photocarriers are swept quickly out of the quantum wells) there is never an appreciable number of carriers in the quantum wells so that $d\sigma_{ow}/dt \approx$ 0 and if it is also true that $\tau_{QW-rec} >> \tau_{out}$ (carriers are swept away before they can recombine) then Eq. 1 reduces to:

$$\frac{\sigma_{\rm QW}}{\tau_{\rm out}} = \frac{I}{h\nu} \left(1 - e^{-\alpha L}\right) \,. \tag{3}$$

Using Eq. 3 in Eq. 2 gives

$$\frac{d\sigma_{d}}{dt} = \frac{I}{hv} (1 - e^{-\alpha L}) - \frac{\sigma_{d}}{\tau_{d,rec}} , \qquad (4)$$

This rate equation is a key result which serves as a starting point for all subsequent calculations.

In the absence of illumination:

$$\frac{d\sigma_d}{dt} = -\frac{\sigma_d}{\tau_{d-rec}}$$
(5)

so that:

$$\tau_{d-rec} = -\frac{\sigma_d}{\frac{d\sigma_d}{dt}}.$$
 (6)

In this experiment, the quantities σ_d and $\frac{d\sigma_d}{dt}$ are determined by measuring the fractional change in transmission $\Delta T/T$ of the n-i-p-i which is produced by the presence of the photocharge. In general, the dependence of the quantity $\Delta T/T$ on σ_d is complicated and often unknown. For this calculation it is assumed that $\Delta T/T$ is proportional to σ_d (in general the dependence is not expected to be linear, but empirically this has been found to be approximately correct) so that:

$$\tau_{d-rec} = -\frac{\Delta T}{T} \frac{d(\Delta T)}{d(\Delta T)}.$$
(7)

Note that this expression can be used to compute an instantaneous lifetime when the lifetime is not constant, as in the case of n-i-p-i structures.

B. Time Resolved Photocarrier Decay - Experiment

For the MQW H nipi sample, transmission was measured through a 2 mm diameter aperture with a weak (= $100 \ \mu$ W/cm²) 853 nm probe beam from a CW dye laser operating with Styryl 9m dye. The transmitted probe was detected with an S-1 photomultiplier and time resolved with a box-car integrator. The sample was excited with 853 nm optical pulses of duration 100 μ sec and intensity 815 mW/cm² at a repetition rate of 0.5 kHz. The excitation pulses were long enough so that the photocarrier population was expected to reach steady state during excitation.

Fig. 2 shows the change in transmission as function of time. This curve was used to determine both $\Delta T/T$ and $d(\Delta T/T)/dt$ for several times after the excitation was



turned off. These values were used in Eq. 7 to determine lifetimes. Because each

Fig. 2. Fractional change in the transmission of a MQW H nipi sample due to an optical pulse.

value for the lifetime has a corresponding $\Delta T/T$, and each $\Delta T/T$ a corresponding intensity (the CW intensity required to produce, in the steady state, that $\Delta T/T$) the dependence of lifetime on intensity can be determined. This dependence is shown in Fig. 3 (open boxes).



Fig. 3. Lifetime in the MQW H nipi as a function of the corresponding intensity. The open boxes are the results of time resolved measurements. The closed circles indicate lifetime inferred from steady state nonlinear absorption spectra.

C. Photocarrier Lifetime From Nonlinear Absorption Spectra - Theory

In this section excess carrier lifetime is deduced from steady state nonlinear absorption spectra. As in sub-section A, the starting point is Eq. 4.

In the steady state, Eq. 1 becomes:

$$0 = \frac{I}{hv} (1 - e^{-\alpha L}) - \frac{\sigma_d}{\tau_{d-rec}}, \qquad (8)$$

which can be solved for τ to give:

$$\tau_{d-\text{rec}} = \frac{\sigma_{d} hv}{I(1-e^{-\alpha}L)}.$$
 (9)

The electric field in the intrinsic region of a n-i-p-i is:

$$\mathsf{E} = \frac{\sigma_0 \cdot \sigma_p}{\varepsilon},\tag{10}$$

where σ_0 is the space charge per unit area in a depleted doped region of the unilluminated n-i-p-i, and ε is the permittivity of the i-region. Equation 10 can be used to eliminate σ_d in Eq. 9:

$$\tau_{d-rec} = \frac{h\nu}{I(1-e^{-\alpha L})} (\sigma_0 - \epsilon E)$$
 (11)

This is the key expression of this sub-section. With Eq. 11, nonlinear absorption spectra can be used to determine the electric field at a given pump intensity and the corresponding lifetime.

To determine a value for the quantity σ_0 to be used in Eq. 11 note that, within the depletion approximation, the thickness w_n of the depleted layer in the n-region of a p-i-n structure with doping p = n is:

$$w_{n} = \frac{Na}{Na+Nd} \left(-w_{i} + w_{i} \left(1 + \frac{2VBI}{w_{i}\gamma}\right)^{1/2}\right), \quad (12)$$

where w_i is the width of the intrinsic region, $\gamma = eN_aN_d/\epsilon(N_a+N_d)$ e is the electronic charge, Na (Nd) is the number of acceptors (donors) in the p-region (n-region), and VBI is the built-in voltage given by:

$$V_{BI} = E_G + kT \ln \left(\frac{N_a N_d}{N_c N_v}\right), \qquad (13)$$

where EG is value of the energy band gap of the material, kT is the thermal energy, and N_C and N_V are the effective density of states in the conduction and valence bands respectively. The quantity σ_0 is computed using Eqs. 12 and 13 together with the relation:

$$\sigma_0 = w_n \times N_d \tag{14}$$

For the MQW H nipi the values of N_a and N_d are both 2 x 10¹⁸, E_G = 1.80 eV is used (the band gap of Al_{0.3}Ga_{0.7}As), and a value of 12.1 is taken for the dielectric constant of the intrins c region. Then w_n = 28 Å and σ_0 = 5.6 x 10¹¹ cm⁻².

D. Photocarrier Lifetime From Nonlinear Absorption Spectra - Experiment

In these experiments, nonlinear transmission of the MQW H nipi was measured, at room temperature, for wavelengths between 650 nm and 930 nm using a microscope lamp as a low intensity broad band probe, the 815 nm CW output of a Styryl 9m dye laser as a pump source, and an optical multichannel analyzer as a detector. The intensity of the probe $(100 \ \mu\text{W/cm}^2)$ was kept as small as possible, while still retaining a reasonable signal to noise ratio, in order to minimize its pumping effects. Fig. 4 shows the change in the absorption coefficient of the quantum wells, as a function of wavelength and pump intensity, deduced from the transmission measurements. The spectral dependence of the changing absorption can be understood in the following way. In the absence of strong illumination the built-in field which appears across the quantum wells is quite strong, which broadens the excitonic absorption resonances and shifts 'hem to lower energies. This is the well known

quantum-confined Stark effect⁴. The pump photo-excites carriers which screen the built-in field. As the excitonic peaks shift to higher energies, the absorption coefficient increases at wavelengths slightly shorter than a resonance while, decreasing at slightly longer wavelengths. Features which correspond to the n=1 and n=2 excitonic resonances are visible in the spectra.



Figure 4. The change in absorption of a MQW H nipi after excitation with a dye laser pump.

In Fig. 4, note that the maximum absorption change (long wavelength dip near 853 nm) shifts to larger energies at larger intensities. We have measured the magnitude of this shift, relative to the energy of the maximum absorption change in the spectrum for a 50 mW/cm², and used the values to infer carrier lifetimes in the following way. The shifts are assumed to be equal in size to the corresponding shift of the n=1 excitonic absorption feature so that published data³ can be used to determine the internal electric fields. The internal fields, together with the corresponding intensities, are used in Eq. 8 to estimate a lifetime.

For this project, the intensity dependence of carrier lifetime, computed using the approach just outlined, has been compared with the values deduced from the time resolved measurements. The results, shown in Fig. 3, indicate good agreement so that more confidence can be placed in the values obtained.

3. LATERAL DIFFUSION EFFECTS IN N-I-P-I STRUCTURES

Lateral diffusion effects are small in the MQW H nipi because the relatively large recombination rates ($t_{d-rec} \approx 10 \ \mu$ sec at large intensities) mean that diffusion lengths ($\approx 100 \ \mu$ m when $t_{d-rec} = 10 \ \mu$ sec) are much smaller than the laser spots (D = 2 mm) used to excite the sample. Nevertheless, diffusion is a potential source of cross-talk in n-i-p-i based devices and chips may have to be pixellated in order to achieve a sufficient density of processing elements. In this study, diffusion effects are characterized in order to understand their contribution to excess carrier lifetime.

A. Lateral Diffusion in Two Dimensional Structures - Theory

The effects of lateral carrier diffusion can be included in Eq. 4:

$$\frac{\partial \sigma_{d}}{\partial t} = \frac{I}{hv} (1 - e^{-\alpha L}) - \frac{\sigma_{d}}{t_{d-rec}} + D \frac{1}{r} \frac{\partial}{\partial r} r \frac{\partial \sigma_{d}}{\partial r}$$
(15)

where D is the ambi-polar diffusion coefficient. If I = 0, then Eq. 15 reduces to:

$$\frac{\partial \sigma_{d}}{\partial t} = -\frac{\sigma_{d}}{t_{d \cdot rec}} + D \frac{1}{r} \frac{\partial}{\partial r} r \frac{\partial \sigma_{d}}{\partial r}$$
(16)

If it is assumed that at t =0 the distribution of photocarriers is Gaussian:

$$\sigma_{d}(t=0) = \sigma_{0} e^{-r^{2}/r_{0}^{2}}$$
(17)

and, for the sake of simplicity, that t_{d-rec} is constant, then for times t > 0:

$$\sigma_{d}(\mathbf{r},t) = \frac{\sigma_{0}r_{0}^{2}}{4Dt + r_{0}^{2}} e^{-t/t_{d-rec}} e^{-r^{2}/(4Dt + r_{0}^{2})}$$
(18)

A generalized lifetime can be defined analogous to Eq. 6:

$$\frac{1}{\tau} = -\frac{\frac{\partial \sigma_{d}}{\partial t}}{\sigma_{d}(t)}$$
(19)

Then, from Eq. 19, it is found that:

$$\frac{1}{\tau} = \frac{1}{\tau_{d-rec}} + \frac{4D}{4Dt + r_0^2} \left(1 - \frac{r^2}{4Dt + r_0^2}\right)$$
(20)

where the second term on the right hand side is the contribution from diffusion. Note that at t=0 and r=0 the value of this term is $4D/r_0^2$. If D is taken to be a typical value of 15 cm² and $r_0 = 0.5$ mm, then, ignoring for the moment τ_{d-rec} , $\tau = 40 \,\mu$ sec. At all other positions r > 0 and times t > 0 the contribution is smaller (the diffusive lifetime is longer).

B. Lateral Diffusion in Two Dimensional Structures - Experiment

Experimental studies of lateral diffusion in a hetero n-i-p-i structure with coupled quantum wells are examined in this sub-section. The coupled quantum well hetero n-i-p-i structure (CQW H nipi)⁴ consists of pairs of 48Å quantum wells separated by narrow 36Å barriers. There are twelve such pairs in each i-region. It was found that, for the range of internal fields that are accessible by optical pumping, the "coupled" wells are actually uncoupled. The structure behaves as if there are 24 uncoupled 48 Å wells per i-region.



Figure 5. A coupled quantum well hetero n-i-p-i structure.

That lateral diffusion of photocarriers is significant in this structure is clearly indicated in Fig. 6. The CQW H nipi structure was excited in its center with a heliumneon laser spot of diameter ≈ 0.5 mm. A low intensity probe was created by passing

the output of a microscope lamp through a monochromator. The probe wavelength of 825 nm was chosen to coincide with the excitonic absorption peak in the quantum wells where the sample transmission depends on the local density of excess carriers. Fig. 6 shows the relative change in the probe transmission as a function of the pump-probe separation. The data can be fit to an exponential with decay constant = 3 mm. The origin of the these exceptionally long diffusion lengths is the corresponding long excess carrier lifetimes. Using L = 3 mm and D = 15 cm²/sec in the expression $\tau = L^2/D$ gives $\tau \approx 200 \mu$ sec. As will be seen, this value is consistent with time resolved measurements of nonlinear transmission. Note a diffusion length of 3 mm implies a minimum pixel size of \approx 6 mm diameter, clearly an unacceptable value.



Figure 6. The nonlinear transmission of the CQW H nipi sample as a function of pump-probe separation. The diagram on the right explains the geometry for measurements.

The effects of carrier diffusion on material response time were examined in more detail. As indicated in the previous sub-section, and particularly in Eq. 20, lateral carrier diffusion can contribute to a local decay rate for photo-carriers.

Fig. 7 shows the lifetime of photocarriers (including both carrier recombination and lateral diffusion) calculated from time resolved nonlinear transmission measurements, similar to those described in sub-section B of section 2, on the CQW H nipi sample using:



Figure 7. Measured lifetime in the CQW H nipi as a function of the corresponding nonlinear transmission for pump intensities of 97 mW/cm² (diamonds) 45 mW/cm² (crosses) and 17 mW/cm² (open circles).

The quantity τ is plotted versus the corresponding fractional change in transmission. Each curve represents a different initial pump intensity. In the simplest picture, ignoring lateral diffusion, there is a one to one relation between $\Delta T/T$ and τ so that the three curves are expected to lie on top of each other. That the curves do not coincide can be attributed to the effects of diffusion. In the case of the CQW H nipi, the recombination lifetime (> 200 µsec) is always longer than the initial diffusive lifetime (= 40 µsec). Thus, immediately after the excitation is removed, the decay of the photocarrier density, within the probed area, is dominated by lateral diffusion. This decay rate, unlike the recombination rate, is roughly independent of the initial pumping intensity. As a result, the lifetime of the right most point on each curve (corresponding to t = 0 when the excitation is just removed) is approximately constant.

4. CARRIER TRAPPING IN THE QUANTUM WELLS OF HETERO N-I-P-I'S

Further measurements have been made which indicate that, even after the effects of lateral diffusion have been minimized by uniformly exciting the sample (see Fig. 6) it is insufficient to describe the decay of photocarriers with a model



Figure 8. Nonlinear transmission of the CQW H nipi under uniform illumination. Subsequent measurements of time resolved nonlinear transmission were made with the probe in the center of sample where the carrier concentration gradient is small and the contribution of diffusion to the carrier decay rate is negligible.

that assumes all carriers decay at the same rate (see Fig. 9). It is likely that some portion of the photocarriers, created in the quantum wells, never escape to the doped regions. These carriers will have relatively short lifetimes. For the case in which the excitation is uniform, the decay of carriers in the quantum wells determines the initial lifetime as measured by time resolved nonlinear transmission.

A two lifetime model has been developed which describes quantitatively the decay of excess carriers in the CQW H n-i-p-i sample. The generalized version of Eq. 4 is:

$$\frac{d\sigma_{tot}}{dt} = \frac{I}{h\nu} (1 - e^{-\alpha L}) - \frac{\sigma_d}{\tau_{d-rec}} - \frac{\sigma_{QW}}{\tau_{QW-rec}}, \qquad (22)$$

where σ_{tot} is the sum of σ_{QW} , the excess carrier density in the quantum well regions, and σ_d , the excess carrier density in the doped regions, and the other quantities have been defined in Section 2. Eq. 22 is used to solve for the steady state densities and



Figure 9. Measured lifetime in the uniformly illuminated CQW H nipi as a function of the corresponding nonlinear transmission for initial pump intensities of 40 mW/cm² (left most curve) 4 mW/cm² (middle) and 0.7 mW/cm² (right most curve). Even though lateral diffusion has been made negligible, the curves do not overlap, so that Eq. 4 can not describe the decay.

subsequently the equation is used to follow the time evolution of the system. The following additional assumptions are made.

1) The two lifetimes depend exponentially on the corresponding space charge potentials:

$$\tau_{d-rec} = \tau_0 e^{(\phi/E_0)}, \qquad \tau_{QW-rec} = \tau_0 e^{(\gamma\phi/E_0)}, \qquad (23)$$

where $\tau_0 = 1$ nsec is of the order of the lifetime for photoexcited carriers in comparable structures with no doping and no built-in electric fields, ϕ is the space charge potential, γ takes into account that only a fraction of the space charge potential separates carriers in the quantum wells, and E₀ is an "activation energy" for the structure. The adjustable parameters γ and E₀ are taken to be 0.5 and 26 meV respectively.

2) The change in the space charge potential is linear in σ_d and σ_{QW} so that:

$$\Delta \phi = \frac{\sigma_d + \eta \sigma_{QW}}{\varepsilon} w, \qquad (24)$$

where w_i is the width of the intrinsic region and η is an adjustable parameter which describes the "effectiveness" of carriers in the quantum wells at screening the built-in fields, taken to be 0.5 for these calculations.

3) As assumed in Section 2, the change in transmission is proportional to the space charge potential.

4) In the steady state, the ratio $\sigma_{QW}/\sigma_d = \Gamma$ is independent of the pumping intensity. The quantity Γ is chosen so as to give the best fit to the experimental data.

Figure 10 shows the calculated carrier lifetimes with $\Gamma = 0.75$. The fit to the data in Fig. 9 is excellent. Note that these results imply that fully 3/4 of the carriers photoexcited in the quantum wells never leave the wells.



Figure 10. Ex as carrier lifetime in the CQW H nipi as a function of the corresponding nonlinear transmission, calculated from the two lifetime model. The initial pump intensities are as in Fig. 9.

5. CONCLUSIONS AND RECOMMENDATIONS

The decay of optical nonlinearities in hetero n-i-p-i structures is determined by the lifetime of photogenerated carriers. Hence, photocarrier lifetime determines the "turn-off" time of optoelectronic devices based on these materials. In this study decay rates have been observed which indicate that, in n-i-p-i samples, photocarriers have a lifetime which increases as the density of photocarriers decreases. In a MQW H nipi sample, the lifetime ranges for \approx 10 µsec to \approx 1 millisecond. Because the decay can not be described by a constant carrier lifetime, it is useful, for device applications, to define a fall time τ_{fall} = the time it takes for a transmission change $\Delta T/T$ to decrease by 50% from the saturated value once the excitation has been removed. For the MQW H n-i-p-i structure, $\tau_{fall} \approx$ 50 µsec.

Hetero n-i-p-i samples must be designed keeping the effects of lateral diffusion in mind. Exceptionally long excess carrier lifetimes can lead to enormous diffusion lengths and cross-talk in nonlinear optical elements spaced even millimeters apart. Structure pixellation can inhibit lateral diffusion but at the expense of very much shorter photocarrier lifetimes, due to large non-radiative recombination at pixel surfaces, and correspondingly larger operating intensities. Passivation of pixel walls by chemical treatment in order to reduce the surface recombination velocity should partially restore the sensitivity of these materials. Alternatively, hetero n-i-p-i structures incorporating Indium could be used as it has been found that these materials have inherently smaller surface recombination velocities. We propose to study the nonlinear optical properties of InAs/GaAs strained layer quantum wells in n-i-p-i structures recently fabricated at the Hughes Research Laboratories.

That a substantial portion of photocarriers may never escape from the quantum wells of a hetero n-i-p-i structure, as demonstrated by studies of the CQW H nipi, also impacts device design. Photocarriers must be swept from the quantum wells in order to screen built-in fields so that carriers trapped in quantum wells are less effective in producing optical nonlinearities. To insure that the greatest possible fraction of carriers tunnel out of the quantum wells, structures should have large built-in electric fields (i-regions as short as possible) and narrow/shallow barriers between wells.

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