Scaling of Stark-Shifted Per-Carrier Nonlinearities in Multiple-Quantum-Well Device Structures

A. N. Cartwright, Student Member, IEEE, X. R. Huang, and Arthur L. Smirl, Senior Member, IEEE

Abstract—We demonstrate simple rules for the scaling of per-carrier Stark-shifted nonlinearities with well number, electric field, amplitude and width of the excitonic transition by measuring the per carrier nonlinear response of a number of multiple-quantum-well structures as a function of temperature, bias and materials system. These measurements illustrate that the per-carrier nonlinearity can be improved by optimizing the in-well bias field and by increasing the number of wells per intrinsic region; however, they also demonstrate that when the measured per-carrier nonlinearities are corrected for material quality and temperature (i.e., excitonic amplitude and linewidth) that the per-carrier response does not depend appreciably on material system or on whether the fields are externally-applied, built-in, or intrinsic.

I. INTRODUCTION

A LARGE optical nonlinearity (absorptive or refractive) per absorbed photon (or injected electron-hole pair) is required for optical switching and logic devices. Device structures (such as self-electrooptic effect devices (SEED’s), hetero n-i-p-i-s, and piezoelectric multiple quantum wells (MQW’s)) that rely on the screening of applied, built-in or intrinsic fields are expected to have large per-carrier nonlinearities primarily because the carriers generated in a single well can escape and move to screen the electric field experienced by all of the quantum wells [1], [2]. As a class, we refer to the nonlinearities in such structures as Stark-shifted nonlinearities, since they arise from a reduction in the quantum-confined Stark effect (QCSE) as the field is screened by the photogenerated charge [3].

Many MQW device configurations, materials, and orientations of materials have been investigated in attempts to improve the nonlinear response of Stark-shifted semiconductor structures. These studies have included device structures in which the electric field was externally applied (e.g., a SEED [4]), built-in (e.g., a hetero n-i-p-i [5, 6]) and intrinsic (e.g., piezoelectric MQW’s [7]). The MQW’s used in these structures were grown under a variety of conditions, were of varying quality, and ranged from lattice-matched materials like GaAs–AlGaAs [8] to strained lattice-mismatched materials like GaAs–InGaAs [9]. The materials used, both strained and unstrained, were grown in a variety of orientations ranging from [100] ([4], [9]) to [111] ([7]). Moreover, experimental parameters, such as optical excitation level, perpendicular field (whether it was applied, built-in or intrinsic) and sample temperature, varied dramatically from study-to-study. For all of these reasons, quantitative comparisons of the nonlinear responses of various Stark-shifted structures have been difficult. First-principle theoretical comparisons are similarly difficult because they depend upon bandstructure and other parameters that are generally not precisely known.

In this paper, we establish a simple phenomenological scaling procedure that allows a systematic comparison of the per-carrier response of Stark-shifted MQW structures. This procedure does not require extensive painstaking fundamental calculations or rely on a detailed knowledge of the band parameters or the materials quality. We establish this scaling by performing an extensive experimental investigation and by presenting a simple phenomenological treatment of the per-carrier optical nonlinearity in a number of hetero n-i-p-i, SEED and piezoelectric MQW structures. We investigate how this figure-of-merit scales with excitonic linewidth, excitonic amplitude, electric field and the number of wells per intrinsic region. These studies include the dependence of the per-carrier nonlinearity on temperature, on materials system and on sample structure. Moreover, they include structures in which the electric fields are externally applied, built-in and intrinsic. We demonstrate that a single simple expression will account for the scaling of the per-carrier nonlinearity with each of the stated parameters. This expression gives us predictive capabilities when designing new structures.

II. A PER-CARRIER FIGURE-OF-MERIT FOR STARK-SHIFTED NONLINEARITIES

For purposes of illustrating and contrasting the definitions and characteristics of bleaching and Stark-shifted per carrier nonlinearities in MQW structures, we limit our discussions to the \( n = 1 \) heavy-hole excitonic transition, and we phenomenologically represent that transition by a Lorentzian [10] or a Gaussian [11] with amplitude \( \alpha_0 \) and a halfwidth at half maximum of \( \Gamma_0 \). The choice of lineshape function depends on the dominant broadening mechanism. Homogeneously broadened systems (e.g., phonon broadened) are best modeled by a Lorentzian line-shape function, and inhomogeneously broadened systems (e.g., systems broadened by fluctuations in well thickness) are more accurately modeled by a Gaussian line-shape function [12].
For nonlinearities arising from bleaching, the nonlinear absorptive cross section is commonly used to quantify the effectiveness of optically-created carriers in changing the absorption coefficient [2, 13–16]. This cross section, \( \sigma_{eh} \), is simply defined as the absorptive change per carrier:

\[
\sigma_{eh} = \frac{\Delta\alpha(\lambda)}{N},
\]

where \( \Delta\alpha \) is the change in the absorption coefficient, and \( N \) is the number density of photoinjected electron-hole pairs. Such a cross section can be defined rigorously and is a constant of the material or the device structure only under a restricted set of conditions. Specifically, the saturation must be sufficiently small to ensure that the change in absorption, \( \Delta\alpha \), is approximately linear in both fluence and carrier density, and the shape of the absorption profile must not change measurably (e.g., it should not be significantly broadened by collisions with the photoexcited carriers.). The change in absorption in a MQW under these conditions is illustrated in Fig. 1(a) for several fluences (where the excitonic bleaching has been represented by a Gaussian whose amplitude has been reduced in direct proportion to the incident fluence). Under these circumstances, the shape, amplitude and position of the bleaching profile per carrier, \( \Delta\alpha/N \), are independent of excitation conditions [as illustrated in Fig. 1(b)], and a true cross section can be defined.

In direct contrast to nonlinearities associated with saturation (or bleaching), a true per-carrier nonlinear cross section cannot be defined for Stark-shifted nonlinearities. Nevertheless, we will demonstrate that, under a restricted set of conditions (analogous to those necessary for defining a bleaching cross section), a useful per-carrier figure-of-merit, \( \sigma_{eh} \), can be defined for Stark-shifting nonlinearities, and we will demonstrate that this figure-of-merit obeys very simple scaling rules. For reasons that will soon be apparent, we choose to use the peak change in absorbance per carrier (per unit area) as a measure of the strength of the nonlinearity in Stark-shifted systems. Under uniform excitation (i.e., when the sample is optically thin and approximately the same density of carriers \( N \) is excited in each well), the peak change in absorbance per carrier per unit area reduces to:

\[
\sigma_{eh} = \frac{\Delta\alpha(\lambda_{peak})N_{w}l_{w}N_{i}}{NN_{w}l_{w}N_{i}} = \frac{\Delta\alpha(\lambda_{peak})}{N},
\]

where \( l_{w} \) is the width of each quantum well, \( N_{w} \) is the total number of wells per intrinsic region, \( N_{i} \) is the total number of intrinsic regions in a sample and the change in absorption coefficient,

\[
\Delta\alpha = \alpha(E_{\perp} - E_{sc}) - \alpha(E_{\perp}),
\]

is that caused by the screening of the perpendicular electric field \( E_{\perp} \) by a space charge field \( E_{sc} \) that is associated with the photo-generated carriers. The quantity \( NN_{w}l_{w}N_{i} \) is easily recognized as the optical thickness of the sample, \( \Delta\alpha(\lambda_{peak})N_{w}l_{w}N_{i} \) is the total absorbance change and \( NN_{w}l_{w}N_{i} \) is the total number of carriers per unit area excited in the sample.

Fig. 1. Phenomenological representation of spectra of the (a) change in absorption coefficient, \( \Delta\alpha \), and (b) the change in absorption coefficient per-carrier, \( \Delta\alpha/N \), as a function of increasing excitation level for an excitonic bleaching nonlinearity in the regime appropriate for defining a cross section (the units for all quantities are arbitrary).

The calculation of \( \Delta\alpha \) is straightforward for both Lorentzian and Gaussian line-shape functions, under the restrictions 1) that the number of carriers that escape the wells and move to screen the field increases linearly with fluence and photoexcited carrier density, 2) that the resulting photo-generated space-charge field \( E_{sc} \) must be small compared to the initial perpendicular field \( E_{\perp} \) (whether it is applied, built-in or intrinsic), and 3) that the area under the exciton absorption line must remain approximately constant (i.e., bleaching is negligible).

The \( \Delta\alpha \) spectra calculated under these circumstances are shown in Fig. 2(a) for several selected excitation levels, assuming a Gaussian absorption profile and assuming that the photogenerated space-charge field opposes the initial perpendicular field. The \( \Delta\alpha \) spectra for Lorentzian lineshapes are qualitatively similar. Notice that each \( \Delta\alpha \) curve consists of a positive peak followed by a negative peak. It is straightforward to show that the zero crossings of the \( \Delta\alpha(\lambda) \) spectra are given by:

\[
\lambda_{c} \approx \lambda(0) + \Delta\lambda(E_{\perp}) - \frac{1}{2} E_{sc} \left. \frac{\partial \Delta\lambda(E)}{\partial E} \right|_{E_{\perp}},
\]

where \( \lambda(0) \) is the center wavelength of the excitonic transition in the absence of a perpendicular field and \( \Delta\lambda(E) \) gives
the shift of this transition with the field. Therefore, \( \lambda(0) + \Delta \lambda(E_\perp) \) is the center wavelength of the exciton in the absence of photoexcitation. In performing this calculation, we have assumed that the shift in wavelength caused by photoexcitation, \( \Delta \lambda(E_\perp - E_{sc}) - \Delta \lambda(E_\perp) \), is small compared to the excitonic linewidth \( \Gamma_0 \), which allows us to expand the shift in wavelength after excitation \( \Delta \lambda(E_\perp - E_{sc}) \) in a Taylor series about \( E_\perp \). Under the same assumptions, the peaks of the \( \Delta \alpha(\lambda) \) spectra can be shown to occur at:

\[
\lambda_p = \lambda_0 \pm \beta \Gamma_0,
\]

where \( \beta \) is a constant which depends on the line-shape function and is given in Table I. Notice that the separation between the two peaks in \( \Delta \alpha(\lambda) \) is directly proportional to the width of the transition, i.e., \( \Delta \lambda_{\text{peak}} = 2\beta \Gamma_0 \).

Using these values for the peak positions, it is then straightforward to calculate the magnitude of the approximately equal peaks in the absorption spectrum:

\[
|\Delta \alpha(\lambda_{\text{peak}})| \approx \left[ \frac{C \alpha_0 E_{sc}}{\Gamma_0} \right] \frac{\partial \Delta \lambda(E)}{\partial E} \bigg|_{E_\perp},
\]

where \( C \) is also a constant that depends on the lineshape function as given in Table I. By taking advantage of the earlier stated restrictions that the exciton should be sufficiently weak that bleaching is negligible and that the number of carriers that escape the wells and move to screen the fields increases linearly with photoexcited carrier density, the photogenerated space charge field can be written as

\[
E_{sc} = \frac{e f N_w l_w N}{\varepsilon},
\]

where \( e \) is the fundamental charge, \( \varepsilon \) is the dielectric constant of the material and \( f \) is the fraction of the photogenerated carrier density that escapes the wells and moves to screen the perpendicular electric field and where the quantity \( N_w l_w N \) is recognized as the total sheet charge density (number per unit area) per intrinsic region.

From these expressions, it is clear that, if we divide each of the curves in Fig. 2(a) by the number density of the photogenerated carriers to obtain \( \Delta \alpha/N \) [as we have done in Fig. 2(b)], each curve will have a constant shape and a constant amplitude, but with a zero crossing that shifts linearly with excitation level. It is this slight shift in \( \Delta \alpha \) with increasing carrier density that precludes our rigorously defining a cross section that is constant for a given wavelength, as we do for bleaching (contrast with Fig. 1).

Under the conditions given here, however, the peak amplitude of \( \Delta \alpha/N \) is a constant and independent of excitation density. It depends only on the materials parameters and is, therefore, a good candidate to serve as a per-carrier figure-of-merit. This is the quantity that we have identified in (2), and it is now evident that it has the following form:

\[
\sigma_{eh} = \pm \frac{e f C N_w l_w \alpha_0}{\varepsilon \Gamma_0} \left. \frac{\partial \Delta \lambda(E)}{\partial E} \right|_{E_\perp}.
\]

Several properties of this expression for the per-carrier figure-of-merit \( \sigma_{eh} \) deserve comment. First, \( \sigma_{eh} \) is proportional to the amplitude of the excitonic transition \( \alpha_0 \) and inversely proportional to the width of the excitonic transition \( \Gamma_0 \). Thus, if the area under the excitonic transition remains constant, then narrower excitonic transitions lead to larger changes in the absorption coefficient per carrier (i.e., larger \( \sigma_{eh} \)'s) but over a smaller wavelength range. Second, it is possible to enhance the nonlinear optical response by increasing the number of quantum wells being screened. Third, in the low field limit regime where \( \Delta \lambda \propto E_\perp^2 \) (i.e., for fields below \( \sim 40 \text{ kV/cm} \) [12], [17], [18]), the per-carrier nonlinearity can be increased by increasing the transverse electric field \( E_\perp \). Finally, \( \sigma_{eh} \) is proportional to the fraction of carriers which contribute to the screening of the perpendicular electric field. In the remainder of this paper we will experimentally verify these properties of the per-carrier figure-of-merit.

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**Table I**

<table>
<thead>
<tr>
<th>Line-Shape Function</th>
<th>Gaussian</th>
<th>Lorentzian</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \alpha_0 )</td>
<td>( \frac{\alpha_0}{\sqrt{\pi}} )</td>
<td>( \frac{\alpha_0}{\pi} )</td>
</tr>
<tr>
<td>( \beta )</td>
<td>( \frac{\beta}{\sqrt{2\ln 2}} )</td>
<td>( \frac{\beta}{\sqrt{\pi}} )</td>
</tr>
<tr>
<td>( \sigma_{eh} )</td>
<td>( \frac{e f C N_w l_w \alpha_0}{\varepsilon \Gamma_0} )</td>
<td>( \frac{e f C N_w l_w \alpha_0}{\varepsilon \Gamma_0} )</td>
</tr>
</tbody>
</table>
TABLE II
CHARACTERISTICS OF THE SAMPLES USED IN THE MEASUREMENTS REPORTED IN THIS PAPER. MATERIAL SYSTEM A IS Al0.15Ga0.85As–GaAs AND MATERIAL SYSTEM B IS GaAs–In0.14Ga0.86As

<table>
<thead>
<tr>
<th>Sample</th>
<th>N_e</th>
<th>N_i</th>
<th>N_w</th>
<th>N_D</th>
<th>d_e = d_w</th>
<th>E_i</th>
<th>E_m</th>
<th>d_t</th>
<th>Material System</th>
</tr>
</thead>
<tbody>
<tr>
<td>W1</td>
<td>1</td>
<td>2</td>
<td>2</td>
<td>20</td>
<td>2</td>
<td>20</td>
<td>77</td>
<td>130</td>
<td>A</td>
</tr>
<tr>
<td>W2</td>
<td>2</td>
<td>1.5</td>
<td>2</td>
<td>20</td>
<td>2</td>
<td>20</td>
<td>77</td>
<td>130</td>
<td>A</td>
</tr>
<tr>
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<td>2</td>
<td>20</td>
<td>2</td>
<td>20</td>
<td>77</td>
<td>130</td>
<td>A</td>
</tr>
<tr>
<td>W3</td>
<td>3</td>
<td>2</td>
<td>2</td>
<td>20</td>
<td>2</td>
<td>20</td>
<td>77</td>
<td>130</td>
<td>A</td>
</tr>
<tr>
<td>W10</td>
<td>10</td>
<td>1</td>
<td>2</td>
<td>20</td>
<td>2</td>
<td>20</td>
<td>77</td>
<td>130</td>
<td>A</td>
</tr>
<tr>
<td>W50</td>
<td>50</td>
<td>1</td>
<td>2</td>
<td>20</td>
<td>2</td>
<td>20</td>
<td>77</td>
<td>130</td>
<td>A</td>
</tr>
</tbody>
</table>

N_e = Number of quantum wells per intrinsic region.
N_i = Number of intrinsic regions.
N_D = Doping density of the n(p) region.
d_e = Width of the n(p) region.
d_t = Width of the intrinsic region.

TABLE III
RESULTS OF MEASUREMENTS ON THE SAMPLES DESCRIBED IN TABLE II. THE MEASURED AND SCALABLE FIGURES OF MERIT ARE GIVEN IN THE LAST TWO COLUMNS, RESPECTIVELY.

<table>
<thead>
<tr>
<th>Sample</th>
<th>T (K)</th>
<th>Applied Bias (V)</th>
<th>( n_e ) (10^8 cm^-3)</th>
<th>( \lambda_0 ) (nm)</th>
<th>( \Gamma_e ) (meV)</th>
<th>( \sigma_e ) (A/cm^2)</th>
<th>( \Delta E ) (meV)</th>
<th>( d_e ) (nm)</th>
<th>( d_t ) (nm)</th>
<th>( N_w ) (10^15 cm^-3)</th>
<th>( N_i ) (10^15 cm^-3)</th>
<th>( d ) (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>W1</td>
<td>300</td>
<td>0.0</td>
<td>13.4</td>
<td>855.4</td>
<td>1.82</td>
<td>3.8</td>
<td>0.48</td>
<td>20</td>
<td>15</td>
<td>50</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td>W2</td>
<td>300</td>
<td>0.0</td>
<td>41.3</td>
<td>855.4</td>
<td>1.82</td>
<td>3.8</td>
<td>0.48</td>
<td>20</td>
<td>15</td>
<td>50</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td>W3</td>
<td>300</td>
<td>0.0</td>
<td>13.5</td>
<td>855.4</td>
<td>1.82</td>
<td>3.8</td>
<td>0.48</td>
<td>20</td>
<td>15</td>
<td>50</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td>W10</td>
<td>300</td>
<td>0.0</td>
<td>13.5</td>
<td>855.4</td>
<td>1.82</td>
<td>3.8</td>
<td>0.48</td>
<td>20</td>
<td>15</td>
<td>50</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td>W50</td>
<td>300</td>
<td>0.0</td>
<td>13.5</td>
<td>855.4</td>
<td>1.82</td>
<td>3.8</td>
<td>0.48</td>
<td>20</td>
<td>15</td>
<td>50</td>
<td>30</td>
<td>15</td>
</tr>
</tbody>
</table>

III. MATERIALS SYSTEMS AND STRUCTURES STUDIED
The details of the seven sample structures in which we measured the per-carrier nonlinearity are given in Table II. These samples included three lattice matched [100]-oriented GaAs–AlGaAs hetero n-i-p-i’s with 1, 2 and 4 wells per intrinsic region (labeled W1, W2, and W4, respectively); a strained [100]-oriented InGaAs–GaAs hetero n-i-p-i with 3 wells in the intrinsic region (labeled W3); a strained [100]-oriented InGaAs–GaAs p-i-n MQW structure with 10 wells (W10) in the intrinsic region and one with 50 wells (W50) in the intrinsic region; and a strained InGaAs piezoelectric MQW p-i-n structure grown in the [111] direction with 10 wells (W10P) in the intrinsic region. The \( \sigma_{eh} \)’s in the p-i-n structures were extracted with and without an externally applied bias. Notice that the samples are numbered in accordance with the number of quantum wells per intrinsic region, i.e., \( \frac{W}{N_w} \) has \( N_w \) quantum wells per intrinsic region. The total number of quantum wells in the device is the product of the number of quantum wells per intrinsic region, \( N_e \), and the number of intrinsic regions in the entire sample structure \( N_i \). Table II also gives the doping densities and widths of the doped regions and the estimated perpendicular electric field in the absence of an externally applied bias. The linear optical transmission spectrum of each sample was measured to ensure that the samples were of high quality and to determine the values of the amplitude \( \alpha_0 \), linewidth \( \Gamma_0 \), and the center wavelength \( \lambda_0 \) of the heavy-hole excitonic transition. The values for these parameters measured under the bias conditions and temperature, \( T \), used to extract the values of \( \sigma_{eh} \) reported in Figs. 5 and 6 of this paper are listed in Table III.

IV. EXPERIMENTAL PROCEDURES
The peak per-carrier change in the absorption coefficient, \( \sigma_{eh} \), of each sample was measured using a two-color pump-probe differential transmission technique. The pump and probe pulses were produced by two independently tunable dye lasers, which were synchronously pumped by the frequency-doubled, compressed pulses from an actively mode-locked Nd:YAG laser. Because of the large variation in the spectral position of the heavy-hole exciton as a function of materials system, temperature and perpendicular field strength, we used several combinations of the following dyes in the two lasers: Styryl 13 with a tuning range of \( \sim 900–980 \) nm, LDS 867 with a tuning range \( \sim 840–930 \) nm and Styryl 9 with a tuning range \( \sim 780–850 \) nm. In each combination, both lasers produced well mode-locked, satellite-free pulses of approximately 2 ps duration. The cavity dumpers for the two dye lasers were synchronized, and the jitter between the pulses from the two lasers was determined by a cross-correlation measurement to be less than 10 ps. The repetition rate for the experiment was always adjusted to ensure almost complete sample recovery between pulses (i.e., in the range 25 kHz–1 MHz).

For the differential transmission measurements, the pump laser was focused onto the sample. The spatial profile of the pump was measured to be Gaussian with a spot size of \( 100 \mu m \) (e^{-1} radius) for measurements on the GaAs–InGaAs samples and \( 50 \mu m \) for the GaAs–AlGaAs samples. For each measurement, the pump was tuned to a fixed wavelength, well above the heavy-hole excitonic resonance, and the fluence was adjusted to inject a known density of carriers, \( N_i \), into the sample. The change in transmission induced by these carriers was then measured as a function of wavelength by tuning a weak time-delayed probe pulse from the second laser across the heavy-hole exciton. For all measurements, the probe radius was adjusted to be \( 1/3 \) of the pump radius to ensure relatively constant excitation over the probe spot. The differential transmission spectra,

\[
\frac{\Delta T}{T} = \frac{(T_{on} - T_{off})}{T_{off}},
\]

where \( T_{on} \) is the transmission of the sample with the pump present, and \( T_{off} \) is the transmission of the sample with the pump absent, were acquired by chopping the pump and probe and by using standard lock-in techniques. The change in absorption coefficient at each wavelength was then extracted from the differential transmission spectra using the expression

\[
\Delta \alpha = -\frac{1}{d} \ln \left( 1 + \frac{\Delta T}{T} \right),
\]

where \( d = l_w N_w N_i \) is the total thickness of the QW’s in the sample.

To ensure that we selected a fixed probe delay such that the Stark-shifted nonlinearity was fully formed, but that negligible recombination had taken place, we studied the differential transmission of each sample as a function of time delay. Although the exact time scales and magnitudes depended on sample type and temperature, the time-resolved differential transmission spectra for each sample were consistent with the following generic picture. Initially, the spectral profiles were
indicative of bleaching (see Fig. 1), indicating that most of the photogenerated carriers were located in the wells. Subsequently, this bleaching profile was replaced by one consistent with a blue shift of the exciton (see Fig. 2), indicating that the carriers had escaped the wells and had drifted to screen the built-in, intrinsic or applied perpendicular field. In each case, this blue shift of the exciton persisted until the spatially-separated carriers began to recombine nonexponentially on much longer time scales [19]. From these studies, we concluded that fixed time delays of 100 ps at room temperature and 800 ps at 80 K were sufficient to guarantee that the screening was complete and that there was no significant recombination in each case.

Once we had used this procedure to select the proper fixed delay, we then measured the peak $\Delta \alpha$ as a function of fluence for each sample to ensure that the amplitude of the Stark-shift varied linearly with fluence over the range used to extract $\sigma_{eh}$. The latter, in turn, ensured that we were operating in a fluence regime where $E_{sc} \ll E_{\perp}$ and where it made sense to define a per carrier figure-of-merit. The optically-induced shifts in the center wavelength of the exciton, $\Delta \lambda(E_{\perp} - E_{sc}) - \Delta \lambda(E_{\perp})$, were less than 0.1 of the excitonic linewidth $\Gamma_0(E_{\perp})$ for all measurements of $\sigma_{eh}$ reported here, consistent with the calculations presented earlier.

Under the restricted set of conditions just described, we then measured $\sigma_{eh}$ for each of the samples. The $\sigma_{eh}$'s in the p-i-n structures were extracted with and without an externally applied bias. For purposes of illustrating this procedure, typical measurements of $\Delta \alpha$ and $\Delta \alpha/N$ for sample W10 are shown in Fig. 3(a) and (b), respectively, for three selected fluences within the linear regime at 80 K. Notice that the observed behavior is almost identical to that predicted by the simple phenomenological model presented earlier and shown in Fig. 2. Most importantly, notice that the shape and size of the $\Delta \alpha/N$ spectra are independent of the excitation fluence within this regime. As a matter of procedure, we used the amplitude of the negative peak to determine $\sigma_{eh}$, since it is farther from the light-hole exciton and from the band edge and, therefore, less likely to be influenced by either.

As a final comment on the experimental procedure, we note that the assumption of uniform excitation implied by (2) is reasonably good for each of the samples used here; however, this is not a strict requirement for obtaining a reliable per carrier response. In the linear regime where it is valid to define a $\sigma_{eh}$, this requirement can be relaxed considerably so long as $\Delta \alpha$ and $N$ are replaced by values for the change in absorption coefficient and the density, respectively, that are averaged over the sample thickness (or equivalently, the total absorbance change is divided by the total number of carriers per unit area).

V. SCALING WITH NUMBER OF QUANTUM WELLS

Equation (8) predicts that the per carrier nonlinearity $\sigma_{eh}$ should scale linearly with the number of quantum wells $N_w$, that is screened by each photogenerated electron-hole pair. (We emphasize that $N_w$ is not the total number of wells in the sample, but the number in each intrinsic region.) The most straightforward test of this dependence is to vary $N_w$ while holding all other parameters constant. Consequently, we initially focus our attention on samples W1, W2, and W4. These three samples are identical, except for the number of wells per intrinsic region, $N_w$. Specifically, each sample is a hetero n-i-p-i, and all three were grown in the same MBE machine, one after the other. As reported in Table I, each of these samples has the same well width, the same total number of wells (32), the same doping levels for the n and p regions, the same width for the intrinsic regions and, therefore, identical fields across the intrinsic regions. The $\Delta \alpha/N$ spectra for these three samples are shown in Fig. 4 for a sample temperature of 300 K. Again, we emphasize that these curves were measured in a fluence regime where the amplitude and shape were independent of fluence [20]. Notice that the values extracted for $\sigma_{eh}$ (i.e. the peak values) are approximately in the ratio of 4:2:1 for W4, W2 and W1, respectively, as predicted by (8). Consequently, we conclude that the per carrier nonlinear figure-of-merit $\sigma_{eh}$ does indeed increase linearly with well number, when the number of wells being screened is small. The question then arises as to whether or not this scaling extends to larger numbers of wells per intrinsic region (e.g., as found in SEED devices).

The $\sigma_{eh}$'s extracted for each of our samples are plotted in Fig. 5 as a function of the number of wells per intrinsic region
in each sample. The solid circles labeled W1, W2 and W4 are the \( \sigma_{\text{ch}} \)'s for the AlGaAs–GaAs hetero n-i-p-i's discussed in the previous paragraph and extracted from Fig. 4. The solid squares labeled W3, W10, and W50 are the \( \sigma_{\text{ch}} \)'s measured at 300 K for the GaAs–InGaAs samples. W3 is a hetero n-i-p-i, and W10 and W50 are simple p-i-(MQW)-n structures. The open square and the open diamond represent \( \sigma_{\text{ch}} \)'s measured at 80 K for sample W10 and for the [111]-oriented piezoelectric p-i-n sample W10P [21], respectively. Note that all of the samples studied had roughly the same well widths, \( \sim 10 \) nm, and except for W10P and W50, roughly equal perpendicular fields in the absence of an applied bias, \( \sim 27 \) kV/cm. In order to simplify our comparisons, an externally applied bias was used to adjust the in-well field of W50 and W10P to the same value.

In summary, then, Fig. 5 presents measurements of \( \sigma_{\text{ch}} \) for two materials systems and two temperatures under conditions of constant well width \( l_w \) and constant perpendicular field \( E_\perp \). If we ignore the measurements at 80 K (the open symbols) for the moment, it is apparent that the unscaled values for \( \sigma_{\text{ch}} \) range over two decades and that within each materials system that \( \sigma_{\text{ch}} \) increases linearly with the number of wells being screened \( N_w \) over the entire range 1–50. It is also apparent that the values for \( \sigma_{\text{ch}} \) are consistently higher for the lattice matched AlGaAs–GaAs samples than for the strained GaAs–InGaAs samples. In the next section, we show that the lower values for strained samples are a consequence of the excitonic broadening in those materials.

VI. SCALING WITH EXCITONIC AMPLITUDE AND LINELWIDTH

To verify that the scaling of \( \sigma_{\text{ch}} \) with excitonic amplitude \( \alpha_0 \) and linewidth \( \Gamma_0 \) is correctly described by (8), we plot the normalized quantity \( \sigma_{\text{ch}} \Gamma_0 / \alpha_0 \) as a function of the number of wells in the intrinsic region of each device, as shown in Fig. 6. The values shown were obtained by using the values for \( \sigma_{\text{ch}} \) given in Fig. 5 and the values for \( \alpha_0 \) and \( \Gamma_0 \) reported in Table III. Remarkably, when corrected for excitonic broadening in this way, all results lie on a single straight line that is linearly proportional to the number of quantum wells per intrinsic region, as predicted by (8), independent of materials composition, device type or temperature and regardless of whether the perpendicular field is built-in, externally-applied, or intrinsic!

VII. SCALING WITH PERPENDICULAR ELECTRIC FIELD

To this point we have established that the per-carrier figure-of-merit \( \sigma_{\text{ch}} \) scales directly with well number \( N_w \) and with the amplitude of the exciton transition \( \alpha_0 \), but inversely with the width of the exciton \( \Gamma_0 \); however we have not determined whether \( \sigma_{\text{ch}} \) scales with field as predicted by our simple phenomenological model. In agreement with previous studies [10], [12], [18], we found that the shift in the center wavelength of the excitons in our samples varied quadratically with the perpendicular field \( \Delta \lambda(\mathbf{E}_\perp) = \gamma E^2_\perp \) for in-well fields below \( \sim 40 \) kV/cm and linearly with the field \( \Delta \lambda(\mathbf{E}_\perp) \propto E_\perp \) for fields above this value, as illustrated in Fig. 7 for the [100]-oriented InGaAs–GaAs p-i-(MQW)-n with 50 wells (W50). Consequently, based on (8), we would expect \( \sigma_{\text{ch}} \) to vary linearly with the field in the low field limit and to be independent of the field at higher fields.

We investigated these tendencies by measuring the \( \sigma_{\text{ch}} \)'s of sample W50 as a function of in-well field. The results for sample W50 at 300 K are shown in Fig. 8. From the results shown in Fig. 8, it is not apparent that \( \sigma_{\text{ch}} \) shows the expected dependencies on in-well field. Specifically, while the per carrier response varies linearly with the perpendicular field in the low field regime, it is not at all apparent that it
is independent of field in the high field regime. However, as shown in Fig. 9, this apparent deviation from ideal behavior in the high field regime is a consequence of the field-induced broadening of the exciton in this regime. Specifically, when we correct for excitonic broadening by plotting the normalized quantity $\sigma_{eh} I_0/\alpha_0$ (see Fig. 9), the result has a linear dependence on field for in-well fields below $\sim 40$ kV/cm and is approximately independent of the field for in-well fields larger than this value. This is exactly the behavior expected from our simple scaling expression, (8)!

VIII. SCALING WITH ALL VARIABLES

In the previous sections, we have confirmed the validity of a simple scaling expression (8) that allows one to predictably adjust for changes in well number and electric field in the design of Stark-shifted nonlinear devices and to predictably correct for operation at other temperatures and to predictably scale for conversions between material systems by correcting for the excitonic amplitude and linewidth. According to these scaling rules [see (8)], we should expect the quantity $\sigma_{eh} I_0/(N_w l_w \alpha_0 \partial \Delta \lambda(E_L)/\partial E)$ to be a constant, independent of material quality, material type, temperature, or bias voltage and independent of whether the fields are externally-applied, built-in, or intrinsic. This quantity when calculated for each of measurements presented in Fig. 6 is given in the last column of Table III (where we have used a value of $\gamma = 2 \times 10^{-10}$ cm$^5$/V$^2$ in calculating the derivative $\partial \Delta \lambda/\partial E$). Notice that this value is indeed remarkably constant ($0.48 \times 10^{-10}$ kV-cm to within $\pm 15\%$), again confirming the validity of the scaling given by (8).

We wish to emphasize that the data given in Table III is truly representative. For example, the same constant value for $\sigma_{eh} I_0/(N_w l_w \alpha_0 \partial \Delta \lambda(E_L)/\partial E)$ is obtained for the measurements as a function of in-well field shown in Fig. 9. In fact, this same constant value is obtained for the wide variety of samples presented here over the full range of bias voltages and temperatures that were investigated. Moreover, this value is consistent with values obtained for $\sigma_{eh} I_0/(N_w l_w \alpha_0 \partial \Delta \lambda(E_L)/\partial E)$ from measurements performed in our laboratory on these and similar samples over a five year period by several individuals using multiple laser systems.

IX. CONCLUSION

Thus, we conclude that the simple phenomenological model presented here scales the relative values of the per carrier optical nonlinearity in Stark-shifted structures to within 20% over a wide range of device types, material systems and experimental conditions. Because of its simple predictable scaling properties, this figure of merit can be very useful in comparing the response of apparently disparate materials or structures, even allowing one to correct (or normalize) for differing growth parameters (e.g., well width and materials quality) and operating conditions (e.g., temperature and bias voltage), as we have shown here. Knowledge of the per-carrier response and its scaling properties can be useful when comparing and investigating the magnitudes and origins of fundamental Stark-shifted nonlinearities. As an example of the latter, we have recently used $\sigma_{eh}$ and its scaling properties to compare and contrast in-well and out-of-well screening in piezoelectric multiple quantum wells [22]. In addition, we have also used the knowledge of the scaling of $\sigma_{eh}$ with perpendicular field and excitonic linewidth to demonstrate that the smaller nonlinear response obtained for [111]-oriented piezoelectric MQW's (in comparison to [100]-oriented MQW's) is partially the result of field-induced excitonic broadening by the huge in-well piezoelectric fields, but is primarily the result of a fundamental or growth-related broadening [23].
The usefulness of $\sigma_{eh}$ as a measure of the nonlinear response is derived in large measure from its independence of the excitonic fluence. To achieve this independence, it was necessary to define $\sigma_{eh}$ in the regime where the excitonic shifts are small and where the screening of the space charge field is far from complete. By comparison, many (but not all) practical nonlinear optical devices require large excitonic shifts. The latter requirement raises the question as to whether $\sigma_{eh}$ is of any relevance for actual device applications. As with any figure of merit, the answer depends upon the device structure and the intended application. We have shown elsewhere [20], for example, that if $\sigma_{eh}$ is increased by increasing the number of wells per intrinsic region then this increase is accompanied by a corresponding reduction in the saturation fluence. In terms of the operation of a device in which linearity is important, this means that sensitivity (large response per carrier) is being traded for dynamic range (i.e., range over which the response is linear). The point is that for actual nonlinear optical device applications, $\sigma_{eh}$ may be relevant, but will usually need to be considered together with other factors or other figures of merit.

As a final comment, we note that many applications and potential applications for Stark-shifted devices are electrooptic, rather than nonlinear optical. It is straightforward to show that the nonlinear absorption change per carrier $\sigma_{eh}$, as defined here, is linearly related to the change in the absorption per charge in applied voltage, in the small signal limit. By inference, then, $\sigma_{eh}$ also contains information about the electrooptic response of the structures studied. Similar comments apply to the usefulness of this information in the design of electrooptic devices as were made concerning the direct use of $\sigma_{eh}$ in the design of nonlinear devices.

REFERENCES


A. N. Cartwright (S’95), photograph and biography not available at the time of publication.

X. R. Huang, photograph and biography not available at the time of publication.

Arthur L. Smirli (S’65–M’73–M’84), photograph and biography not available at the time of publication.