In-well screening nonlinearities in piezoelectric multiple quantum wells

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The contribution of the in-well screening of the piezoelectric field to the nonlinear optical response of a [111]-oriented strained InGaAs/GaAs multiple quantum well is separated from, and compared to, that associated with the out-of-well screening and with bleaching by using picosecond nondegenerate differential transmission techniques. These measurements demonstrate that the per-carrier nonlinearity associated with the in-well screening is smaller than that associated with the out-of-well screening by a factor roughly equal to the number of wells. © 1995 American Institute of Physics.

Strained multiple quantum wells (MQWs) that are grown on [111]-oriented substrates have huge intrinsic piezoelectric fields along the growth direction. The presence of this piezoelectric field shifts the exciton to the red through the quantum-confined Stark effect (QCSE). The nonlinearity in these materials then arises as the photogenerated charge screens the piezoelectric field causing the exciton to shift to the blue as the QCSE is reduced. Early theoretical work predicted a large nonlinear response for such materials because of the screening of these piezoelectric fields by the photoexcited carriers that remained in the quantum wells (in-well screening).

To date, however, there have been few measurements of the nonlinear response of piezoelectric MQWs, and all of them have been performed in mechanically clamped structures (e.g., InGaAs/GaAs grown on GaAs). In such structures, the strain and, therefore, the piezoelectric field are localized in the well, and as a result, a potential accumulates across the MQW region. For times long compared to the carrier escape time (~10 ps at room temperature), we have shown that the nonlinearity response of such MQWs is dominated by the long range (out-of-well) screening of the piezoelectric field by photogenerated carriers that have escaped the wells and moved to the edges of the MQW region. Moreover, as part of these studies, we have also demonstrated that in-well screening contributes to the nonlinear response on times short compared to the carrier escape time, but our limited time resolution precluded the quantification of the contributions of in-well screening. To the best of our knowledge, the nonlinearities associated with in-well screening have never been measured quantitatively or compared to the nonlinearities associated with bleaching or out-of-well screening.

In this letter, we definitively isolate the contributions of in-well screening from those of bleaching and out-of-well screening in a [111]-oriented In$_{0.15}$Ga$_{0.85}$As/GaAs MQW, and we compare these three nonlinearities on a per carrier basis. To accomplish this, we ensured that the carriers remained in the wells long enough to measure the contributions of in-well screening by cooling the sample to 25 K. We then performed nondegenerate pump-probe (differential transmission) measurements using two synchronized independently tunable picosecond dye lasers operating at a low repetition rate (25 kHz).

The sample studied here is the same p-i-(MQW)-n structure that was used in our recent work. The band structure of this sample is discussed in more detail elsewhere, but the key features are shown schematically in Fig. 1 for convenience. Specifically, the MQW region consists of ten 10 nm wide In$_{0.15}$Ga$_{0.85}$As/GaAs wells separated by 15 nm wide GaAs barriers. The MQW region is clad by 117.5 nm wide undoped GaAs spacer layers. The sample was grown on an n$^+$ doped [111]-oriented substrate and was capped by a 300 nm thick layer of p$^+$-doped GaAs. The concentration of both n and p dopants (>2×10$^{18}$ cm$^{-3}$) is more than sufficient to allow a built-in potential of ~1.4 V to develop between the doped regions.

The piezoelectric field in the quantum wells is in opposition to the p-i-n electric field, and therefore, both the total in-well field (~100 kV/cm) and the accumulated potential across the MQW region are reduced. The latter reduces the recombination lifetime and ensures that the sample recovers between laser pulses. In fact, the thicknesses of the well and the barrier layers are such that the accumulated decrease in potential over the MQW region due to the piezoelectric field...
is greater than the accumulated increase due to the \( p-i-n \) field. Consequently, there are local potential minima for electrons and holes at opposite ends of the MQW region as shown in Fig. 1.

The nondegenerate differential transmission measurements were performed using two independently tunable, cavity-dumped, cw-modelocked dye lasers. Each laser produced pulses of \( \sim 2 \) ps in duration (full width at half maximum), and there was a measured jitter of \( \sim 10 \) ps between the pulses from the two lasers. One laser was used as a fixed wavelength pump and was set slightly to the blue of the heavy-hole exciton, while the other was used as a variable wavelength probe and was tuned across the excitonic absorption profile. Standard lock-in techniques were used to measure the difference in the probe transmission, \( \Delta T \), with and without the pump present as a function of probe wavelength, pump fluence, and delay time between pump and probe pulses. The differential absorption spectrum was then extracted from differential transmission using the expression: 

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

where \( L \) is the total thickness of the quantum wells.

The nonlinear response and carrier dynamics are illustrated in Fig. 2. The absorption spectra measured at selected time delays following excitation are shown in Fig. 2(a) and the concurrent blueshifts of the exciton extracted from a more complete set of measurements are shown in Fig. 2(b). Notice that immediately following excitation, the excitonic peak is bleached significantly, indicating that most of the carriers are still in the wells. This bleaching is accompanied by an instantaneous blue shift of the exciton, indicating the presence of in-well screening. The bleaching significantly recovers over the next \( \sim 800 \) ps as the carriers escape the wells and move to the edge of the MQW region to screen the accumulated potential and to flatten the bands. This out-of-well screening causes a gradual increase in the blue shift over the same time period. The dynamics portrayed in Fig. 2 are similar to those that we have recently reported for the same sample at a temperature of 80 K, except that the carrier escape was much faster at the higher temperature and prevented measurement of the per-carrier nonlinear in-well response.

Having identified in-well screening and having established the carrier escape time, we then measured the per carrier nonlinear response at a time delay of 20 ps while most of the carriers remained in the wells and at 800 ps when most had escaped. We did this by reducing the pump fluence significantly (compared to that used to obtain the data shown in Fig. 2) and by measuring the bleaching and the blue shift in a regime where these quantities varied linearly with carrier density and where it is appropriate to define per carrier nonlinearities. Under these conditions, the bleaching and the blue shift will be small, and the spectra of the change in absorption coefficient per carrier \( \Delta \alpha(\lambda)/N \), where \( N \) is the carrier density] will have a constant amplitude and shape, although the zero crossings will shift slightly with excitation level. In this regime, the peak change in absorption coefficient per carrier, \( \Delta \alpha_{\text{eh}} \), will be independent of carrier density and can be taken as a valid per-carrier figure of merit.

The results of measuring \( \Delta \alpha(\lambda)/N \) under these conditions at a time delay of 20 ps (while most of the carriers remained in the wells) are shown in Fig. 3(a). By attributing the observed reduction in excitonic amplitude to bleaching and the observed blue shift to in-well screening, this spectrum can be readily separated into two components, as illustrated in Fig. 3(b). The bleaching component (dashed curve) was obtained by numerically shifting the blue-shifted and bleached spectrum after excitation \( \alpha(\lambda,20 \text{ ps}) \) back to the center wavelength of the unexcited exciton and subtracting it from the unexcited spectrum \( \alpha(\lambda,-20 \text{ ps}) \). The in-well screening component (solid curve) was then obtained by subtracting the shifted and unshifted versions of \( \alpha(\lambda,20 \text{ ps}) \).

Note that, in the limit in which \( \Delta \alpha_{\text{eh}} \) is defined (i.e., the blue shift and the bleaching are small compared to the excitonic wavelength and amplitude, respectively), approximately the same result is obtained by first blue shifting the unexcited spectrum to obtain the in-well screening component, then subtracting the shifted unexcited spectrum from the excited spectrum to obtain the bleaching component. That is, in the small signal limit, the peak magnitudes of the components are relatively insensitive to the assumed order of the decomposition.

The \( \Delta \alpha(\lambda)/N \) spectrum measured at a time delay of 800 ps after most (but not all) of the carriers had escaped the wells and moved to the local minima is shown in Fig. 3(c). However, because the carrier escape was not entirely complete after 800 ps at 25 K (the maximum time delay available without changing the experimental configuration), we raised the carrier temperature to 80 K to reduce the carrier escape time to \( <100 \) ps, and we repeated the measurement of \( \Delta \alpha(\lambda)/N \) at 800 ps. Under conditions of complete escape,
the maximum per carrier change $\sigma_{eh}$ (measured at the negative peak) increased from $\sim 18 \times 10^{-14}$ cm$^2$ [as shown in Fig. 3(c)] to $\sim 30 \times 10^{-14}$ cm$^2$. We have separately shown$^9$ that the per carrier response $\sigma_{eh}$ increases with decreasing temperature (because the exciton narrows and increases in amplitude with decreasing temperature). Therefore, $\sim 30 \times 10^{-14}$ cm$^2$ measured at 80 K represents a conservative estimate of the value that would be obtained at 25 K after the carriers fully escape the wells.

From these measurements, it is clear that we have been able to separate the contributions of in-well screening in piezoelectric MQWs from those of bleaching and those of out-of-well screening. Moreover, the per carrier nonlinearity associated with in-well screening $[\sigma_{eh}(\text{in-well}) \sim 2 \times 10^{-14}$ cm$^2]$ is much smaller than that associated with out-of-well screening $[\sigma_{eh}(\text{out-of-well}) \sim 30 \times 10^{-14}$ cm$^2]$ and also smaller than that associated with bleaching $[\sigma_{eh}(\text{bleaching}) \sim 4 \times 10^{-14}$ cm$^2]$—not an order of magnitude larger$^4$ or comparable$^2$ as originally suggested. These relative values are, however, in excellent agreement with more recent calculations.$^{10}$

The principal reason that the out-of-well per-carrier screening response is larger than the in-well screening response (by a factor $\gg$ well number) is that, when carriers occupy a single well, they can screen only the field in that well (and then only partially). By contrast, when the carriers escape that well and move to the edge of the sample, they can simultaneously screen the fields in all of the wells$^{11}$ (in this case 10). Also, notice that the presence of bleaching tends to obscure and further reduce the effectiveness of the in-well screening. The absolute and relative strengths of the per carrier responses associated with bleaching and in-well screening will, of course, depend upon charge separation, and therefore, well width. For a given in-well field, the per-carrier response associated with in-well screening (bleaching) would be expected$^9,10,12$ to decrease (increase) with decreasing well width (assuming that the wells are narrow enough to produce confinement and that the MQW structure is homogeneously broadened).

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