Effect of confinement on energy-dependent dephasing in heterostructures

S. R. Bolton,* S. Bar-Ad, G. Sucha,† and D. S. Chemla
Department of Physics, University of California, Berkeley, California 94720
and Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720

D. L. Sivco and A. Y. Cho
Bell Laboratories, Lucent Technologies, 700 Mountain Avenue, Murray Hill, New Jersey 07974
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To study the effects of confinement by quantum-well potential discontinuities on excitonic dephasing, we performed a spectrally and temporally resolved study of band-edge four wave mixing emission from a series of $In_{1-x}Ga_xAs$ quantum wells. Our measurements reveal an array of dynamics as we move from the three-dimensional to the two-dimensional limit. Spectral resolution allows us to resolve a slowly dephasing excitonic contribution in bulk $In_{1-x}Ga_xAs$. In measurements on quantum-well samples of intermediate width, we find no change of the dephasing time as the quantum-well width becomes smaller than the bulk Bohr diameter. This indicates that the dominant dephasing mechanism in this regime is scattering by alloy disorder and interface roughness. For quantum-well widths below 200 Å we observe a substantial increase of the dephasing time. Spectral resolution allows us to associate the slow dephasing in this regime with localized excitons. [S0163-1829(97)04424-X]

I. INTRODUCTION

Ultrafast dynamics in semiconductors have been studied with great vigor ever since the development of ultrafast laser sources in the early 1980s. The effects of quantum confinement on these dynamics pose important questions both scientifically, for the understanding of fundamental condensed matter interactions, and technologically, for the development of electro-optic devices. The physics which underlies the answers to these questions, however, is not well understood. It is known that most of the many body interactions important in ultrafast dynamics, such as screening, Pauli blocking, and the carrier-phonon interaction, are different in two dimensions than in three. However, real structures exist in a regime intermediate between the absolute two- and three-dimensional limits, and in this regime very little theoretical or experimental work has been done. The ultrafast dynamics of real semiconductors generally result from a complex interplay among many different physical processes. Because of this complexity, it is very difficult to model exactly how confinement will affect the ultrafast properties of a particular structure. Previous work has measured the effects of confinement in semiconductor heterostructures on linear optical properties.[1] Studies of carrier thermalization[2] in such heterostructures have found that confinement substantially increases the effectiveness of Pauli blocking nonlinearities at the band edge, while reducing the effectiveness of the electron-phonon interaction for nonthermal electron populations. A study of the effect of confinement on spin dynamics was also reported recently.[3]

Polarization relaxation, or dephasing, takes place via the same types of collisions which are important in carrier thermalization. These include carrier-carrier scattering, carrier-phonon scattering, and carrier-impurity scattering. Polarization relaxation, however, reflects elastic collisions, which only cause a change of phase, as well as inelastic ones, which also result in a redistribution of energy. At the band edge, where photocarriers cannot lose excess kinetic energy, elastic scattering dominates. Studies of the excitonic dephasing as a function of dimensionality are thus necessary to complement energy and spin relaxation measurements.

Previous measurements of dephasing in confined systems have shown that the dephasing time tends to increase as a result of confinement. This has been shown for confinement in a quantum well (QW), as well as for confinement by a magnetic field.[4] This study, however, explored only the two extreme cases of material confinement: narrow quantum wells and bulk. From these two limits alone it is not clear what mechanism dominates the increase of dephasing time in narrow QWs. Bulk materials and narrow quantum wells clearly represent drastically different scattering regimes. In the strong confinement regime exciton localization plays an important role.[5] Exciton localization is due to lateral confinement by well-width fluctuations, and has been shown to result in a substantial suppression of dephasing.[6] Previous work, however, does not allow separation of the effect of localization from that of confinement by the QW. In this context it is important to realize that transverse confinement by the QW potential and lateral confinement (localization) do not necessarily take place at identical QW widths. At the bulk limit, interpretation of existing experimental data is complicated by the fact that continuum states as well as excitonic states are simultaneously excited by any ultrashort pulse. The emission is thus composed of contributions from different species, for the strongly confined, as well as the intermediate and bulk regimes. Therefore spectral resolution of the excitonic emission is vital for understanding the mechanisms which bring about dephasing. In addition, no previous work has explored the evolution from two to three dimensions as a function of QW width.

In order to address these issues, we have performed a spectrally and temporally resolved study of band-edge four

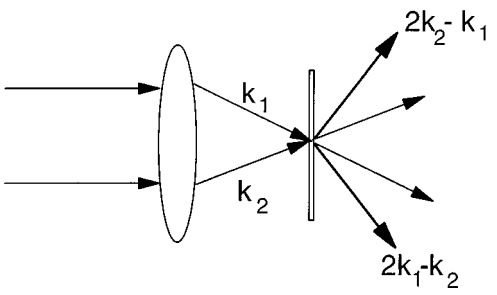

FIG. 1. Schematic FWM geometry. The directions to which the coherent emission occurs are shown in bold.

wave mixing (FWM) emission from a series of $In_{1-x}Ga_xAs$ QWs. The well widths of our samples range from 6000 to 100 Å, and thus cover a wide range compared to the bulk exciton Bohr radius (290 Å).

In Sec. II of the paper we review the application of transient FWM to the study of dephasing in semiconductors. We discuss the limitations of the basic four wave mixing technique, and how confinement in increasingly narrowing QWs is expected to affect scattering and dephasing, thereby demonstrating how the use of spectral resolution and the right choice of samples can help in elucidating the complicated interplay of various physical effects. Section III briefly describes the experimental setup. The experimental results are then presented and discussed in Sec. IV, while conclusions are drawn in Sec. V.

II. DEPHASING MECHANISMS AND MEASURING THE EFFECTS OF CONFINEMENT

A. Transient four wave mixing in semiconductors

Transient FWM measurements are performed using two noncollinear pulses which are time delayed from one another. The two pulses, propagating with wave vectors k_1 and k_2, generate a third-order polarization and result in emission into the momentum conserving directions $2k_1-k_2$ and $2k_2-k_1$ (Fig. 1). In the simplest form of the experiment, a slow detector is used to collect the diffracted photons, and the resulting FWM signal is measured as a function of the delay between the two excitation pulses. In this configuration, positive delay is defined for emission in direction $2k_1-k_2$ $(2k_2-k_1)$ as the case when the pulse with wave vector k_2 (k_1) is exciting the sample first.

In the past ten years, transient FWM has been exploited to gain access to information about coherent dynamics in various optically excited media. In the case where the laser excites a system of uncoupled two-level dipoles,[7] the FWM signal is emitted at positive delays only. Its intensity decays exponentially as a function of time delay with a characteristic time $T_2/2$ for a homogeneously broadened system, or $T_2/4$ for an inhomogeneously broadened one, where T_2 is the dephasing time of the ensemble. A number of studies, however, have shown that a simple exponential decay is the exception rather than the rule in semiconductors. This is due primarily to the fact that a semiconductor cannot be described as an ensemble of independent two-level systems. In many cases, several different resonances are excited by the pump beam (for example, both light and heavy hole excitons), leading to the appearance of quantum beats in the emitted FWM signal.[8] Even in the case where a single excitonic level is excited, Coulomb correlations substantially modify the FWM signal, leading to a nonexponential decay and emission at negative time delays.[9,10] These effects substantially complicate the interpretation of FWM signals in terms of dephasing dynamics. In order to extract more accurate information than is available in a simple FWM experiment, we have made spectrally resolved measurements of the FWM emission as a function of time delay between pulses. In the case where several resonances are emitting simultaneously, spectral resolution allows for interpretation of nonexponential decays. In addition, FWM line shapes can be used to understand interactions in the system, such as Coulomb coupling among states.[11,12]

B. Scattering, dephasing, and the effects of confinement

Many scattering processes can influence dephasing dynamics. Early work[13] demonstrated a strong temperature dependence in excitonic FWM decay times, indicating the importance of exciton-phonon scattering. At low (10 K) temperatures, however, the phonon population is quite small and exciton-phonon scattering is rather slow. Other strong dephasing mechanisms are exciton-electron and exciton-hole scattering. These can be minimized by using low-density excitation and by tuning the excitation so that very little energy is available to excite free carrier states above the exciton. This is feasible mainly for narrow QW samples, where the excitons are strongly bound. In the case of wide QW and bulk samples, which do not show excitonic resonances which are clearly separated from the continuum, exciton-electron and exciton-hole scattering may be important at all densities. For high-density excitation, where the excitonic resonances are nearly saturated, production of unbound electrons and holes again becomes important even in narrow QWs. When the presence of free carriers is minimized, exciton-exciton interaction has been shown to strongly affect FWM emission.[9–11,13] Schultheis *et al.* have shown that at low temperatures and for low excitation densities, exciton-exciton scattering can dominate the dephasing process in QWs.[13] Finally, scattering by charged impurities should also be considered as a dephasing mechanism for the low-temperature, low-density case.

We now focus on the effects of confinement on dephasing. It has been shown that when excitons are reduced in spatial extent, dephasing times tend to lengthen. In one study of dephasing times in 100 Å $In_{1-x}Ga_xAs$ QWs and $In_{1-x}Ga_{As}$ bulk,[4] the dephasing time was six times longer in the QWs. Studies of both bulk and QWs showed that when excitons are reduced in spatial extent by the application of a magnetic field, their dephasing times also tend to increase.[4] This increase in dephasing time with decreasing spatial extent of the exciton has several possible origins. The first is a suppression of scattering by impurities and alloy disorder. As excitons are reduced in size, they become less likely to encounter and thus scatter from these imperfections. How the increase in dephasing time should scale with exciton diameter, however, is not clear. Another possible explanation for the increase in dephasing time would be a decrease in exciton-exciton scattering. Finally, the effect of scattering by

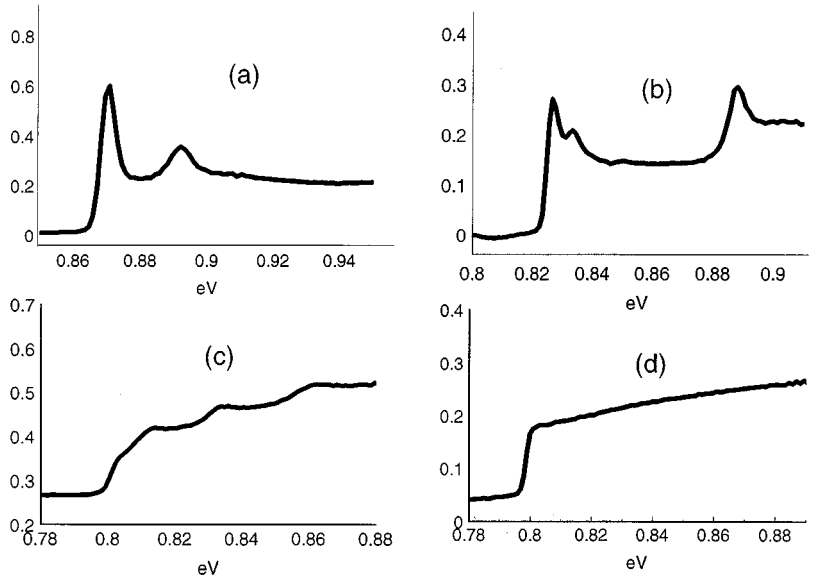

FIG. 2. Band-edge absorption spectra for (a) 100 Å, (b) 200 Å, (c) 500 Å, and (d) 6000 Å samples.

free carriers cannot be ruled out as an explanation for the short dephasing times measured in the three-dimensional (3D) limit.

For the low-temperature case, in the low-density limit, one would expect any dimensionality dependence of the dephasing time to be dominated by changes in the exciton interaction with material defects. There are two opposing contributions to this effect. As discussed above, the decrease in exciton cross section with confinement would be expected to increase the excitonic dephasing time, for the case of a fixed impurity concentration. However, as one goes to very narrow quantum wells, where the well width is substantially smaller than the exciton diameter, the importance of scattering from defects at the interfaces will increase. Although for the infinite barrier case, the excitonic wave function has a node at the interfaces, and thus excitons should not directly ''contact'' the interfaces, charged impurities at the interface will affect the excitons over long distances. Furthermore, for finite barriers, the electron and hole wave functions do leak somewhat into the barrier layers, and this leakage is known to increase with decreasing well width. Thus scattering due to interface roughness should increase with increasing confinement in a QW potential. On the other hand, interface roughness can actually result in a *smaller* scattering cross section: it is known to result in an inhomogeneously broadened absorption line with a mobility edge at its center.[6] Localized excitons, below the mobility edge, can become delocalized by thermal activation. As the temperature is decreased, the lowest-energy excitons become virtually immune to activation, resulting in a significant increase of their dephasing time. Depending on details of sample quality, then, confinement could either increase or reduce excitonic dephasing times.

The limited experimental data in the literature[4] do not allow a detailed analysis of confinement effects in real samples in terms of the scattering mechanisms discussed above. In the following we address the above issues by presenting, for the first time, a detailed study on a series of $In_{1-x}Ga_xAs$ QW samples, ranging in well width from 100 to 6000 Å. The use of $In_{1-x}Ga_xAs$ material has the advantage (over GaAs QWs) that the 2D limit is reached at a larger QW width (due to the larger Bohr radius of the exciton and the larger confinement potential), decreasing the significance of exciton localization. In fact, the transition from 3D to 2D, around 600 Å (one Bohr diameter), is well separated from the localization regime for typical experimental conditions. This is because at liquid He temperature the fluctuations in the ground state energy of a 200 Å QW, due to monolayer fluctuations, become comparable to k_BT. Therefore we expect localization effects to set in at well widths below $\approx$200 Å, well below the transition from 3D to 2D in $In_{1-x}Ga_xAs$ (note that the change of ground state energy due to a monolayer change of the QW width L scales as L^{-3}). In the ternary $In_{1-x}Ga_xAs$ material, alloy fluctuations may also contribute to both scattering and exciton localization. However, we do not expect this relatively well width-independent property to change the sharp onset of localization by monolayer fluctuations. Importantly, spectrally resolving the FWM signal allows us to separate the different contributions within the inhomogeneously broadened absorption line associated with localization. Furthermore, it also enables us to separate the different contributions from continuum states and excitonic states, which are simultaneously excited in wide QWs and in bulk material.

III. EXPERIMENTAL TECHNIQUE

Spectrally resolved FWM experiments were performed on $In_{0.47}Ga_{0.53}As$ heterostructures with well widths of 100, 200, 500, and 6000 Å. The excitonic binding energy in these samples ranges approximately from 2.5 meV (bulk) to 10 meV (100 Å well). All the samples contain a total of 6000 Å of $In_{0.47}Ga_{0.53}As$, with $In_{0.47}Al_{0.53}As$ barriers of 70 Å. The semi-insulating InP substrates were removed by selective etching to eliminate the background two-photon absorption contribution to the signal.[14] All measurements reported here were carried out at liquid He temperature. Absorption spectra of the samples at 4 K are shown in Fig. 2. Note that the excitonic linewidths (or the rise of the band edge, in the case of the bulk sample) are very similar, about 8 meV full width at half maximum (FWHM), for all samples. Thus judging from linear data alone one would expect that the

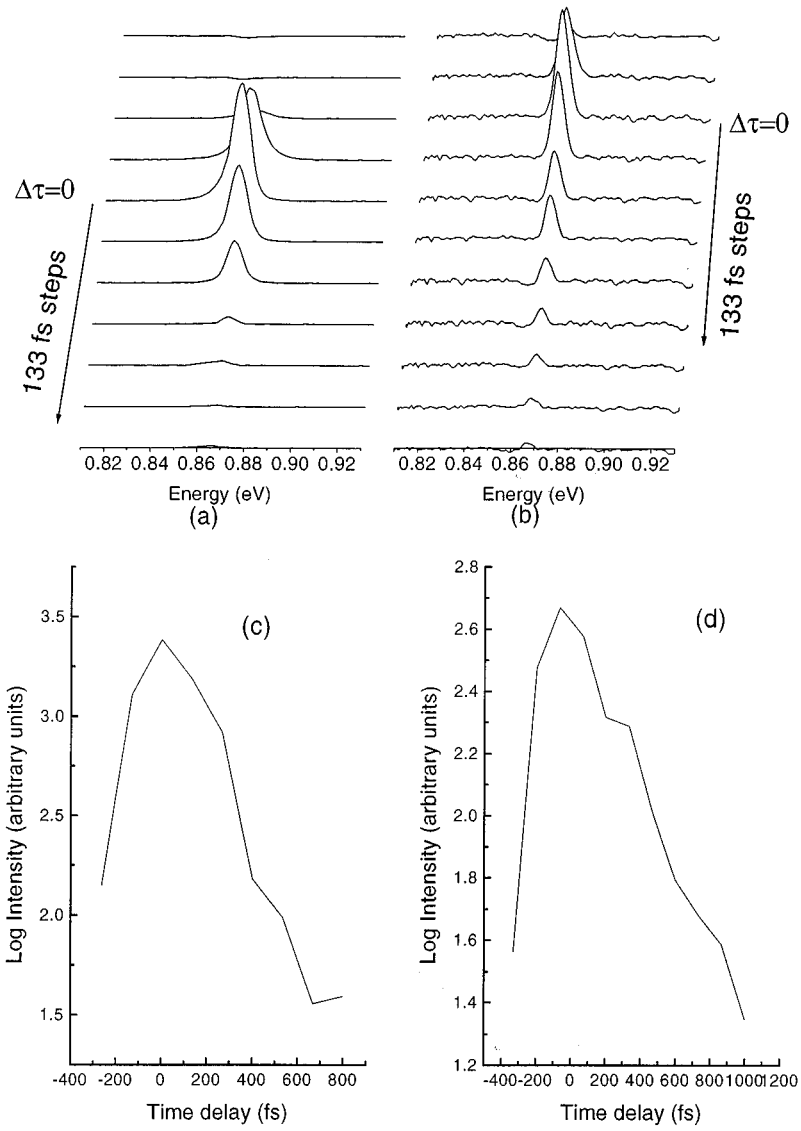

FIG. 3. FWM signal from the 100 Å sample: (a) power spectra as a function of time delay for $N=10^{11}$ cm^{-2}; (b) power spectra as a function of time delay for $N=10^{9}$ cm^{-2}; (c) spectrally integrated FWM as a function of time delay for $N=10^{11}$ cm^{-2}; (d) spectrally integrated FWM as a function of time delay for $N=10^{9}$ cm^{-2}.

excitonic interactions are also similar among all samples.

Samples were excited with 120 fs pulses of approximately 30 nm full width half maximum bandwidth. These were selected with interference filters from the continuum generated by a NaCl laser, amplifier, and continuum generation system.[15] The excitation was tuned just below the lowest exciton level of each sample, in order to minimize the creation of free electron-hole pairs. The measurements were performed over densities ranging from 10^{15} to 10^{17} cm^{-3} (10^{9} to 10^{11} cm^{-2}). The two exciting beams were colinearly polarized and of approximately equal intensity. The signal in the background-free, FWM direction was sent to a spectrometer and $In_{1-x}Ga_xAs$ optical multichannel analyzer (OMA) and measured as a function of time delay between pulses. For each sample, measurements were also performed with a spectrally integrating detector. However, due to the low repetition rate (1 kHz) of the system, averaging of the OMA signal gave a considerably better signal to noise ratio than did the combination of an integrating detector and a lock-in amplifier, allowing for measurements at considerably lower densities. Thus data shown here are all obtained from the spectrally resolved measurements, which can of course be integrated to obtain the total emitted intensity as a function of time delay. We will discuss data from each of the samples in turn, starting with the narrowest quantum well, which has in many respects the simplest response.

IV. EXPERIMENTAL RESULTS

A. Two-dimensional regime

The 100 Å QW has its 1s heavy hole exciton peaked at 870 meV. This sample was excited with 15 meV (FWHM) pulses centered around 880 meV. The results of spectrally resolved FWM are shown in Fig. 3(a) and Fig. 3(b) for both high- (10^{11} cm^{-2}) and low- (10^{9} cm^{-2}) density excitation. Figures 3(c) and 3(d) show the spectrally integrated signal, which gives exponential decays with a time constant of 325 fs for low-density excitation, and 190 fs for the high-density case. If we were to interpret the spectrally integrated signal only, assuming that the excitonic line in this case is inhomogeneously broadened, then the two-level system result for this sample would correspond to dephasing times T_2 of 750 fs and 1.3 ps for the high and low densities, respectively. The

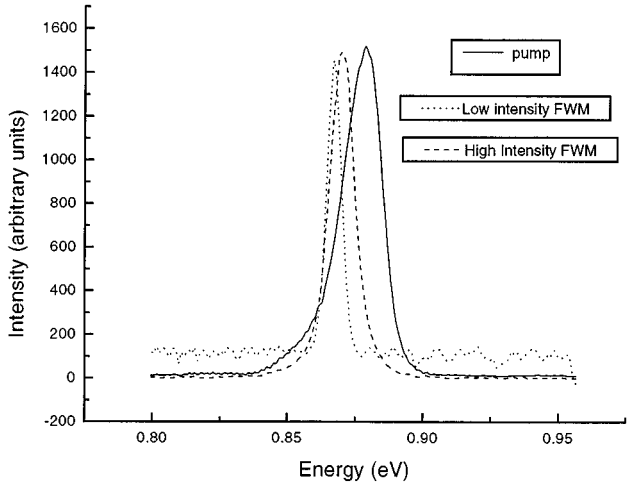

FIG. 4. FWM spectra measured with the 100 Å sample at $N=10^9$ cm^{-2} (dotted) and $N=10^{11}$ cm^{-2} (dashed) overlaid with the pump spectrum (solid).

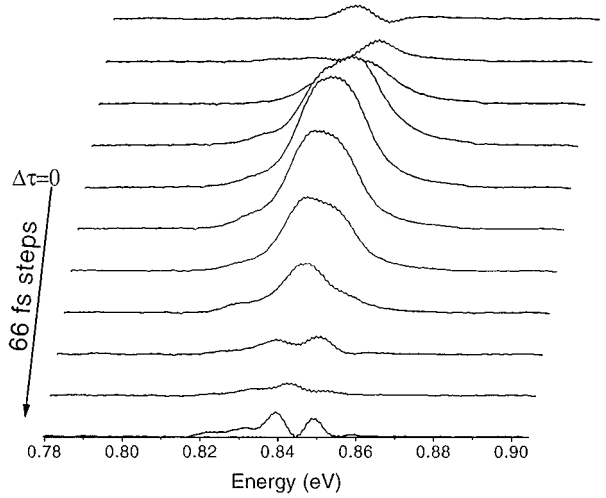

FIG. 5. FWM power spectra as a function of time delay measured with the 200 Å sample at a density $N=10^{10}$ cm^{-2}.

assumption of inhomogeneous broadening in this case is quite reasonable, due to well-width inhomogeneity and alloy fluctuations, as discussed above. If we take 1.3 ps as a lower limit for the dephasing time, we obtain an upper limit for the excitonic homogeneous linewidth of approximately 3 meV. The absorption spectrum for this sample (Fig. 2), however, shows a linewidth of approximately 8 meV (FWHM), thus indicating that the sample is, in fact, quite substantially inhomogeneously broadened. It is likely, in fact, that if we were able to make FWM measurements at lower densities we would obtain even longer dephasing times. Further information is revealed by a closer examination of the FWM spectra for the high- and low-density cases. Figure 4 shows the FWM spectra at high and low densities for $\Delta t=0$, overlaid with the pump spectrum. Note that with increasing density, the emission both broadens considerably and shifts towards the pump spectrum and the peak of the exciton absorption line (Fig. 2). Such an increase in breadth of the emission at high densities has been seen before in GaAs,[11] and has been attributed to the fact that at 10^{11} cm^{-2} the exciton is nearly saturated, and thus the excitation generates a substantial free carrier population. This would sharply decrease the dephasing time and shift the signal from pure excitonic emission to exciton plus free carrier emission. This interpretation conflicts, however, with the symmetrical FWM line shape. Alternatively, the shift and broadening of the response with increasing density may be due to the fact that the lowest lying excitons are localized. At low excitation densities the emission may then be dominated by these localized states, which have longer dephasing times and therefore a stronger integrated emission. The latter interpretation is supported by the fact that the emission spectrum at low density is narrower than the linear absorption line. Surprisingly, we find that the FWM line shape at high-density excitation is well fit by a Lorentzian of ≈ 10 meV FWHM. Conversely, at low density a Lorentzian shape results in a poor fit, while a Gaussian of ≈ 5 meV FWHM gives a much better fit. This means that (a) even the narrow emission line is inhomogeneous and (b) at high densities collision-induced dephasing affects all localized and delocalized excitons in the same way, resulting in a homogeneous emission line. This behavior further underlines the importance of disorder effects in the interpretation of the data from the 100 Å sample.

B. Intermediate regime

The intermediate dimensionality samples (200 and 500 Å) show behavior similar to that in the narrowest well, in that they have dephasing times which increase strongly with decreasing density. In these samples background scattering was a problem, and it was not possible to go to densities below 10^{10} cm^{-2}. Figures 5 and 6 show the spectrally resolved signals for each sample. We obtain a dephasing time T_2 for the 200 Å sample of 530 fs at 10^{10} cm^{-2}, considerably shorter than that measured in the 100 Å sample at the same density. The shorter dephasing time in the 200 Å QW may be due to (a) reduced confinement or (b) reduced localization due to well-width fluctuations. As explained above, the latter is expected to result in increased dephasing times for well widths below 200 Å, but at the same time should manifest

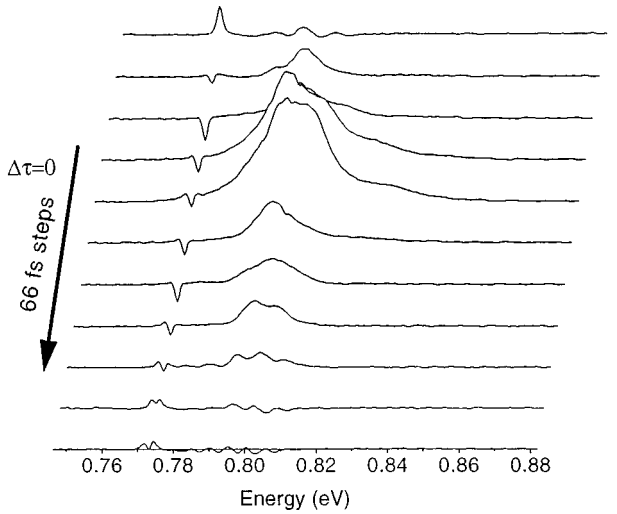

FIG. 6. FWM power spectra as a function of time delay measured with the 500 Å sample at a density $N=10^{10}$ cm^{-2}.

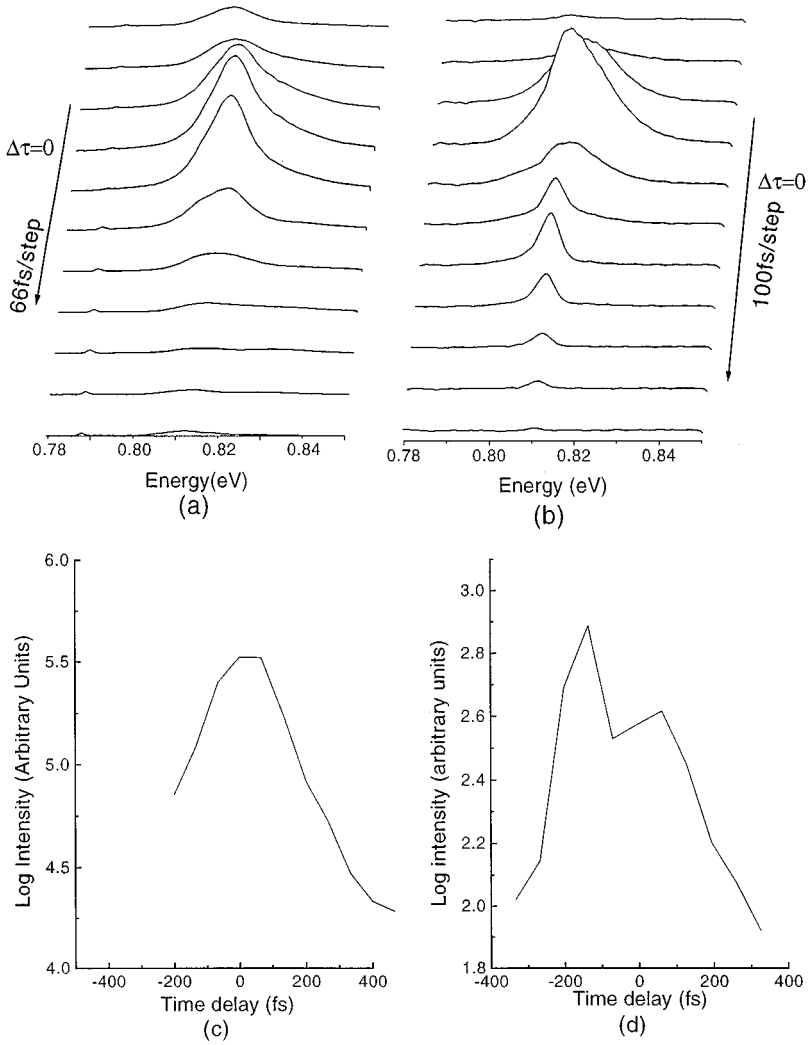

FIG. 7. FWM signal from the 6000 Å sample: (a) power spectra as a function of time delay for $N=3\times10^{16}$ cm^{-3}; (b) power spectra as a function of time delay for $N=1\times10^{15}$ cm^{-3}; (c) spectrally integrated FWM as a function of time delay for $N=3\times10^{16}$ cm^{-3}; (d) spectrally integrated FWM as a function of time delay for $N=1\times10^{15}$ cm^{-3}.

itself as an increasing exciton linewidth. Surprisingly, the linear absorption spectra for these two samples (in fact *all* our samples) show similar linewidths. Moreover, well-width fluctuations cannot account for the ≈ 8 meV linewidths of all but the 100 Å sample. This suggests that a completely different mechanism determines the relatively well-width-independent exciton broadening in our samples. A chief candidate is alloy disorder.

Alloy broadening scales as the square root of the ratio of the unit cell volume to the exciton volume. Therefore in the 2D limit it is expected to increase as both the QW width and the Bohr radius decrease.[16] Conversely, it is expected to be independent of the well width for wide QWs. However, the well-width dependence in the $2d$ limit will be substantially reduced by leakage of the electron and hole wave functions into the barrier layers in the narrow QWs. To demonstrate that alloy fluctuations can in fact account for the experimental linewidths, we estimate the broadening in the 2D limit,[16] assuming a random alloy within the volume of the exciton. The calculated linewidth for the 200 Å sample is ≈ 6 meV, in good agreement with our data.

It is not clear if alloy disorder can, in turn, lead to localization. The answer to this question obviously depends on the length scale of the disorder. In this context it is worth noting that the linewidths seen in the absorption spectra represent averaging over a very large spot size (these measurements are done using incandescent light bulbs), unlike our FWM measurements. In order to answer these questions the data from the 500 Å QW and bulk samples have to be analyzed.

The spectrally resolved signal in the 500 Å sample is shown in Fig. 6. We attribute the highly asymmetrical line shape to the presence of several very closely spaced resonances near the band edge of this sample. The heavy hole and light hole $1s$ excitons are just barely resolvable from one another in the linear absorption. Although we do not resolve separate peaks from these two resonances in the FWM spectrum, we do observe a wide, non-Gaussian, emission spectrum which does not coincide with the excitation spectrum. The FWM power spectra for the 500 Å sample also show a small shoulder around zero delay. We interpret this as a fast decaying emission from the continuum states.

Apart from the above mentioned differences, the FWM emission from the 200 and 500 Å samples shows a remarkable similarity. The dephasing rates and emission linewidths are comparable, and both are very different from those which characterize the 100 Å sample. In spite of the fact that confinement is expected to occur in this regime, as the well width becomes narrower than the bulk Bohr diameter, we do not observe any significant change of the dephasing time.

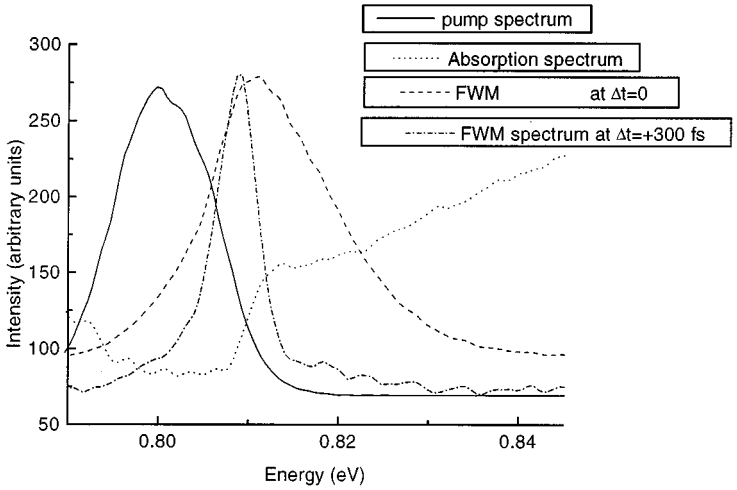

FIG. 8. Spectra measured with the 6000 Å sample: absorption spectrum (dotted), pump spectrum (solid), FWM at $\Delta t=0$ and $N=10^{15}$ cm^{-3} (dashed), and FWM at $\Delta t=300$ fs and the same density (dash-dot).

This is even more surprising considering the decreasing excitation of continuum states as the well width is decreased from 500 to 200 Å. These results can only be explained by assuming a dominant role for scattering by alloy fluctuations and interface roughness. Thus in this regime we observe emission from delocalized excitons, which are scattered by disorder. In turn, this implies that the significant increase in dephasing time in the 100 Å QW sample is due to localization by monolayer well-width fluctuations.

C. Three-dimensional regime

The FWM signals from the bulk sample taken at high and low density are shown in Fig. 7. In this sample we see a qualitative change in the FWM spectrum as we decrease the carrier density. At high densities (above 10^{17} cm^{-3}) the emission is very broad and decays quickly, within 100 fs. As the density is lowered, however, a narrow peak emerges which has a much longer decay time. The broad emission is centered near $\Delta t=0$, while the narrow emission appears to be somewhat time delayed, peaking at approximately $\Delta t=150$ fs. Figure 8 shows the narrow (late time) and broad (early time) signals superimposed on the linear absorption spectrum. The narrow line does not correspond to any feature observed in the linear absorption spectrum. If the sample is excited somewhat above the band edge, then only the broad response is seen. We interpret this in terms of two types of emission. The broad, instantaneous emission derives from the continuum of unbound states near the band edge. The long-lived, narrow line is due to the 1*s* excitonic bound state, which is not resolved from the continuum in linear absorption. Remarkably, the decay time of the narrow line ($\approx$200 fs) is similar to that found for the 100 Å QW ($\approx$325 fs). The resolution of a narrow excitonic feature from a broadband continuum by the difference in dephasing time is the first such measurement to our knowledge. It explains in a very satisfying way the presence of apparently resonant signals in ac Stark effect and carrier thermalization data in this sample,[2] despite the absence of any clear exciton in the linear absorption spectra.

In view of the known sensitivity of excitonic dephasing to scattering by free carriers, it is surprising to find a rather slow excitonic dephasing simultaneous with a clearly resolved free carrier contribution. This, and the fact that the narrow emission line is redshifted relative to the broad exciton absorption peak, suggest that the long-living component is associated with excitons which are localized by alloy disorder. On the other hand, our data show that the fast dephasing in bulk material, which was observed in spectrally integrated measurements,[4] can in fact be attributed to coherent emission by continuum *and* bound states rather than to excitonic emission only.

V. CONCLUSIONS

Table I summarizes the spectral features which we observe in the FWM emission from the four samples, and the typical decay times of the main spectral components, at high and low excitation densities. An array of dynamics is revealed as we move from the three-dimensional to the two-dimensional limit. Rather than obtaining phase relaxation times which evolve smoothly with confinement, we find that

TABLE I. Summary of the spectral line shapes and of the decay times of the main spectral components observed in the FWM emission from the four samples. The accuracy of the high-density data is limited by the pulse duration in all but the 100 Å sample.

	High density		Low density	
Sample	τ (fs)	Line shape	τ (fs)	Line shape
100 Å	190	Lorentzian	325	Gaussian
200 Å	66	Lorentzian	132	Gaussian
500 Å	66	Asymmetric	132	Instantaneous shoulder
6000 Å	66	Asymmetric	200	Long-lived component

for each confinement regime (bulk, intermediate, quasi 2D) unique features in the FWM signal emerge, reflecting different properties of the exciton.

Spectral resolution allows us to resolve a slowly dephasing excitonic contribution in bulk $In_{1-x}Ga_xAs$. It also allows association of the initial fast dephasing with emission by continuum states. In measurements on QW samples of intermediate width, we find no change of the dephasing time as the QW width becomes smaller than the bulk Bohr diameter. This is despite the increasing confinement and a decreasing contribution of free carriers (to both emission and scattering). This indicates that the dominant dephasing mechanism in this regime is scattering by alloy disorder and interface roughness. For a QW width of 100 Å, fluctuations in the ground state energy, due to interface roughness, become larger than the thermal activation energy k_BT. At this point the dephasing time increases substantially, and the FWM spectrum shows clear signatures of localization.

We conclude that scattering and localization due to interface roughness strongly affect excitonic dephasing in QWs. We attribute the different exciton dephasing times for bulk and narrow QWs, reported in the literature, to the fast dephasing of continuum emission and to exciton localization, respectively. The main effect of confinement by the QW potential is therefore a change in the character and role of material defects. This is in contrast with confinement by a magnetic field,[4] which, in the presence of *fixed* material defects, results in a continuous increase of the dephasing time.

Our work also shows the power of nonlinear spectroscopy, in this case allowing discrimination between a long-lived bound state and a continuum, not possible with linear spectroscopy.

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*Permanent address: Department of Physics, Williams College, Williamstown, MA 01267.

†Permanent address: IMRA America, 1044 Woodridge Avenue, Ann Arbor, MI 48105.

[1] W. Stolz, J.C. Maan, M. Altarelli, L. Tapfer, and K. Ploog, Phys. Rev. B **36**, 4301 (1987).

[2] G. Sucha, S.R. Bolton, D.S. Chemla, D.L. Sivco and A.Y. Cho, Appl. Phys. Lett. **65**, 1486 (1994); S.R. Bolton *et al.* (unpublished).

[3] I. Brener, W.H. Knox, K.W. Goossen, and J.E. Cunningham, Phys. Rev. Lett. **70**, 319 (1993).

[4] T. Rappen, G. Mohs, and M. Wegener, Appl. Phys. Lett. **63**, 1222 (1993).

[5] C. Weisbuch, R. Dingle, A.C. Gossard, and W. Wiegmann, Solid State Commun. **38**, 709 (1981).

[6] J. Hegarty, L. Goldner, and M.D. Sturge, Phys. Rev. B **30**, 7346 (1984); J. Hegarty, K. Tai, and W.T. Tsang, *ibid.* **38**, 7843 (1988).

[7] T. Yajima and Y. Taira, J. Phys. Soc. Jpn. **47**, 1620 (1979).

[8] E.O. Gobel, K. Leo, T.C. Damen, J. Shah, S. Schmitt-Rink, W. Schäfer, J.F. Muller, and K. Kohler, Phys. Rev. Lett. **64**, 1801 (1990).

[9] S. Weiss, M.-A. Mycek, J.-Y. Bigot, S. Schmitt-Rink, and D.S. Chemla, Phys. Rev. Lett. **69**, 2685 (1992); M.-A. Mycek, J.-Y. Bigot, S. Weiss, S. Schmitt-Rink, and D.S. Chemla, Appl. Phys. Lett. **60**, 2666 (1992).

[10] K. Leo, M. Wegener, J. Shah, D.S. Chemla, E.O. Gobel, T.C. Damen, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. Lett. **65**, 1340 (1990); M. Wegener, D.S. Chemla, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. A **42**, 5675 (1990).

[11] J.-Y. Bigot, M.-A. Mycek, S. Weiss, R.G. Ulbrich, and D.S. Chemla, Phys. Rev. Lett. **70**, 3307 (1993).

[12] U. Siegner, M.-A. Mycek, S. Glutsch, and D.S. Chemla, Phys. Rev. B **51**, 4953 (1995).

[13] L. Schultheis, J. Kuhl, A. Honold, and C.W. Tu, Phys. Rev. Lett. **57**, 1635 (1986).

[14] S. Bar-Ad, S.R. Bolton, and D.S. Chemla (unpublished).

[15] G. Sucha, S.R. Bolton, and D.S. Chemla, IEEE J. Quantum Electron. **QE-28**, 2163 (1992); G. Sucha, and D.S. Chemla, Opt. Lett. **15**, 1177 (1991).

[16] P.M. Young, E. Runge, M. Ziegler, and H. Ehrenreich, Phys. Rev. B **49**, 7424 (1994).