PHYSICAL REVIEW B VOLUME 38, NUMBER 11 15 OCTOBER 1988-I

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Enhanced inelastic scattering and localization of excitons in $\mathrm{In_{0.53}Ga_{0.47}As/InP}$ alloy quantum wells

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(Received 24 June 1988)

Spectral hole-burning in $\mathrm{In_{0.53}Ga_{0.47}As/InP}$ alloy quantum wells show strong localization of all excitons at low temperatures and enhanced inelastic scattering over GaAs quantum wells. The observed scattering rate agrees with the phonon-assisted tunneling model proposed by Takagahara.

In two-dimensional $\mathrm{GaAs/Ga_xAl_{1-x}As}$ quantum-well structures heavy-hole excitons have exhibited strong inhomogeneous broadening at low temperatures by interface roughness.[1] The roughness can scatter mobile excitons elastically and provides a range in energy for exciton states among which inelastic scattering can occur by phonons. At low temperatures a sharp changeover in energy at the center of the inhomogeneous exciton line has been observed between strongly localized states and states delocalized over a scale of 1 μm.[2] The inelastic scattering rates were found to depend strongly on whether the excitons were localized or mobile, being an order of magnitude greater for the latter. Mobile excitons can additionally scatter off the potential fluctuations. The scattering rates and hence the localization properties are expected to depend strongly on the amount of disorder. Photon echo measurements on quantum wells with extremely flat interfaces,[3] and hence a small amount of disorder, yielded elastic scattering rates consistent with all excitons being mobile. The opposite effect of increased disorder should lead eventually to localization of all states. This has not been measured in these systems to date.

In this paper, we present results on the effects of increased disorder, introduced by alloying the quantum wells, on the localization properties. In $\mathrm{In_{0.53}Ga_{0.47}As/InP}$ quantum-well alloy structures we use a picosecond hole-burning technique[4] to study the exciton dynamics. We find that inelastic scattering is the dominant scattering leading to localization of all excitons to the lowest temperature measured. The scattering rate, however, is found to be much stronger than for localized excitons in GaAs wells and is similar, instead, to the scattering rate of mobile excitons in those wells. The rate is consistent with predictions of Takagahara based on phonon-assisted tunneling of localized excitons.[5]

The samples consisted of 50 layers of $\mathrm{In_{0.53}Ga_{0.47}As}$, 80 Å thick, sandwiched between 100-Å-thick InP layers, grown by chemical beam epitaxy[6] on InP substrates. The heavy-hole exciton line is well resolved in absorption at 5 K with a width [full width at half maximum (FWHM)] of 8 meV.[7] The width is a result of alloy and interface fluctuations together with any layer-to-layer variations at a macroscopic level. Hole burning in the inhomogeneously broadened heavy-hole exciton line in GaAs wells has been previously observed and is a result of saturation of that part of the line resonant with a narrow-band laser.[4] The selective saturation is attributed to filling of the layer space available to excitons resonant with the laser which can occur at a low enough density as to only weakly affect the nonresonant exciton states. The hole was burned with a tunable color center laser mode locked to give 10-psec pulses with a spectral width of 2 Å. The hole was probed in a novel way using a probe beam with the same pulse length but tunable over 2 nm.[8] Tunability was obtained by passing a split-off part of the laser output through 200 m of single-mode silica fiber with sufficient intensity to produce high-order solitons. At an input peak power of 15 W the transmitted beam was spectrally broadened to 2 nm. The fiber output was passed through a spectrometer to give a tunable probe centered at the pump wavelength and with a pulse length similar to that of the probe. Pump and probe beams were cross polarized to minimize any coherent artifacts.

Figure 1 shows a series of spectra of $\Delta T/T$ for different delays after the pump pulse with a photon energy 2.9 meV lower than the peak of the inhomogeneous line. All spec-

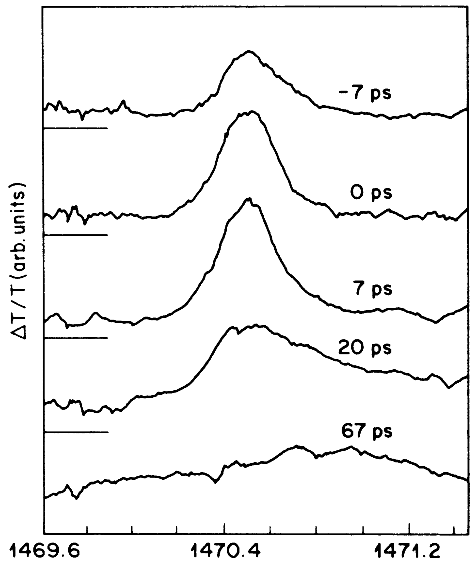

FIG. 1. Fractional change in probe transmission $\Delta T/T$ as a function of wavelength for different delays after the pump. The pump is at 1470.5 nm on the lower-energy side of the heavy-hole absorption peak.

tra show a broad positive background which grows with delay and which is due to saturation of the whole inhomogeneous line by exciton-exciton interaction. On top of the broad background at small delays is a narrow feature resonant with the pump wavelength whose strength initially is 3 to 4 times that of the background. We rule out the possibility that this feature is a coherent artifact[9] by its intensity. A coherent artifact with cross-polarized beams is expected to have the same strength as the signal at finite pump-probe delays. The occurrence of this spectral hole clearly shows that the interaction between resonant excitons is more efficient than between nonresonant excitons. The width of the hole is 3 Å, which is a convolution of pump width, spectrometer resolution, and homogeneous width Γ_h of the hole. The combination of pump and spectrometer widths was measured to be also close to 3 Å so that Γ_h is less than 1 Å. The precise magnitude of Γ_h could not be measured directly. The amplitude of the hole dies away rapidly with time and is zero after about 50 psec.

The exact lifetime of the hole was measured by the ordinary pump-probe method, in which pump and probe had identical spectral profiles. This was achieved by reducing the input intensity to the fiber below the onset of any nonlinearity. The time evolution of $\Delta T/T$ is shown in Fig. 2. The signal decreases rapidly at first, in accordance with the disappearance of the hole, leaving a long-lived component at later times. The long-lived component is exponential with a lifetime of about 2 nsec, close to the fluorescence lifetime of the excitons. It corresponds to the decay of the broad background which at low exciton densities is expected to scale with the total exciton population.

The fast decay of the hole on a time scale much shorter than the exciton recombination time shows that the initially excited population of excitons spectrally diffuse to other states within the inhomogeneous line. A simple rate-equation calculation was performed to fit the data in Fig. 2. The fitting parameters included an exponential spectral relaxation time T_s and exciton absorption saturation densities N_{s1} and N_{s2} for probe photon energy resonant and nonresonant with the pump, respectively. The results are superimposed as the dashed curves in Fig. 2. Since the fitting is sensitive to these parameters, the spectral relaxation time T_s can be determined rather accurately and was found to be 16 psec. The ratio N_{s1}/N_{s2} of ~ 4 was used in the fitting, in agreement with the data of Fig. 1.

The spectral relaxation time T_s varies little with pump wavelength and is also independent of intensity, indicating that we are in the low-density limit. The spectral diffusion is consequently not by exciton-exciton interaction but by inelastic scattering off phonons. A resonant population decay time of 16 psec would set a lower limit of 0.7 Å (0.04 meV) to Γ_h. Since the upper limit to Γ_h is less than 1 Å from Fig. 1, we conclude that the dominant contribution to Γ_h is inelastic scattering off phonons. If the excitons were mobile we would expect greater elastic scattering than in GaAs wells because of the extra alloy scattering contribution. The elastic scattering time for mobile excitons in GaAs wells was measured to be $\simeq 1$ psec, so we conclude that all excitons in $In_{0.53}Ga_{0.47}As$ wells are localized and hop between localized sites incoherently by phonon emission or absorption.

Takagahara[5] has calculated the contributions to the total scattering rate for both localized and delocalized excitons. For localized excitons in the absence of a mobility edge acoustic phonon-assisted tunneling leading to spectral relaxation was the only scattering mechanism considered. This process depends on the exciton density of states, the exciton-phonon coupling coefficient, the phonon density of states, and the temperature. The tunneling rates $1/T_s$ are given by

$$T_s^{-1} = (T_s^0)^{-1}\exp(BT^{\alpha}) ,$$

where T_s^0 and B are constants and α is expected to be in the range 1.6–1.8. In Fig. 3 the measured variation of the scattering rate T_s^{-1} with temperature is shown. The data can easily be fit by the tunneling equation provided

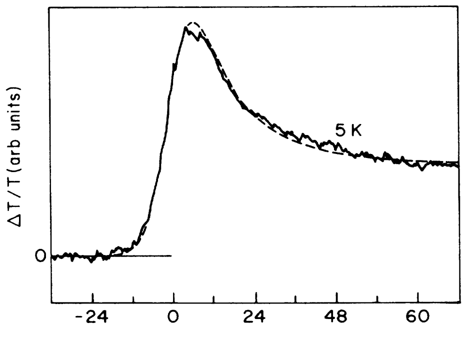

FIG. 2. Pump-probe decay where the pump and probe have the same spectral profiles. The dotted line is a fit to the data using the procedure described in the text. The wavelength is 1470.5 nm. The deconvoluted spectral decay time is 16 ps.

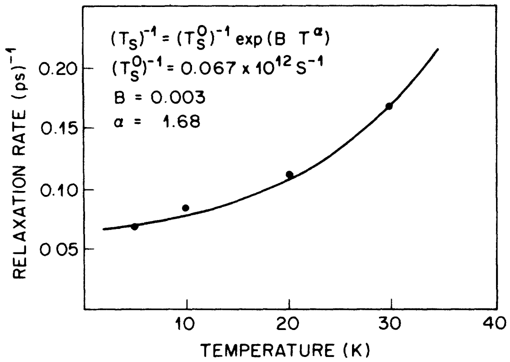

FIG. 3. Plot of the measured spectral relaxation rate T_s^{-1} (points) against temperature along with a fit by the phonon-assisted tunneling model. The wavelength is as in Fig. 1.

$\alpha = 1.68$, as shown in Fig. 3. The agreement of the exponent with that predicted shows that phonon-assisted tunneling is the dominant scattering mechanism. The absolute values of the scattering also agree closely with Takagahara's predictions.

The measured value for Γ_h is only very weakly dependent on position within the inhomogeneous line at the temperatures used. A weak dependence would indicate a tail in the density of exciton states allowing relaxation even at the lowest temperatures and at the lowest measurable energies in the line.

It is interesting to compare the scattering rates with those observed in GaAs wells. The inelastic scattering time in GaAs, 50 Å wide, was as long as 200 psec at $T = 5$ K, much longer than that expected from phonon-assisted tunneling. The inelastic scattering time for the observed mobile excitons, on the other hand, is comparable to that for localized excitons in $In_{0.53}Ga_{0.47}As$. The large difference in inelastic scattering rates for localized excitons may be attributed to a much stronger coupling of the excitons to acoustic phonons in $In_{0.53}Ga_{0.47}As$. The strength of the interaction with LO phonons has already been found to be twice stronger in $In_{0.53}Ga_{0.47}As$ than in GaAs wells.[7]

In conclusion, we have seen that scattering of excitons in $In_{0.53}Ga_{0.47}As/InP$ alloy quantum wells is predominantly inelastic to the lowest temperatures measured. All excitons are consequently in the strongly localized regime and tunnel incoherently from site to site. The scattering rate agrees with Takagahara's predictions but is an order of magnitude greater than in the nonalloy GaAs quantum wells. It should now be possible to fine tune this family of materials between the nonalloy and alloy extremes to determine the cause for the enhanced scattering in the alloy material.

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

[2] J. Hegarty, L. Goldner, and M. D. Sturge, Phys. Rev. B **30**, 7346 (1984).

[3] L. Schulteis, A. Honold, J. Kuhl, K. Kohler, and C. W. Tu, Phys. Rev. B **34**, 9027 (1986).

[4] J. Hegarty and M. D. Sturge, in *Proceedings of the International Conference on Luminescence,* edited by W. M. Yen and J. C. Wright (North-Holland, Amsterdam, 1984), p. 494.

[5] T. Takagahara, Phys. Rev. B **32**, 7013 (1985); J. Lumin. **381**, 64 (1987).

[6] W. T. Tsang, Appl. Phys. Lett. **45**, 1234 (1984).

[7] K. Tai, J. Hegarty, and W. T. Tsang, Appl. Phys. Lett. **51**, 152 (1987).

[8] K. Tai and J. Hegarty (unpublished).

[9] T. Heinz, S. L. Palfrey, and K. B. Eisenthal, Opt. Lett. **9**, 359 (1984).