# Effects of hole burning on pulse propagation in GaAs quantum wells

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(Received 28 December 1981; revised manuscript received 16 February 1982)

Pulse time-of-flight measurements at the heavy exciton resonance in GaAs quantum wells reveal large pulse delays which are dependent on intensity and pulse spectral width. The measured pulse velocities are comparable to those seen in bulk GaAs and are as low as $6.7\times10^6$ cm $s^{-1}$. We obtain quantitative agreement with a simple theory based on hole burning observed in the inhomogeneous exciton line, and we show that the measurements are useful in estimating exciton damping, under certain conditions.

In the linear regime the propagation of short optical pulses in direct-gap semiconductors is described by the dielectric response function $\epsilon(\omega)$ where

$$\frac{\epsilon(\omega)}{\epsilon_\infty}=1+\frac{\Lambda^2}{\omega_0^2-\omega^2-i\omega\Gamma+Dk^2} \quad . \tag{1}$$

$\omega_0$ is the resonant frequency, $\Gamma$ is the damping, $Dk^2$ is a term describing the spatial dispersion of the excitons, $\Lambda^2$ is a measure of the exciton-photon coupling, and $\epsilon_\infty$ is the background dielectric constant. Two regimes are of particular interest here. Firstly, we consider the case of $Dk^2=0$. The frequency dispersion in the real part $n$ of the index of refraction $\sqrt{\epsilon(\omega)}$ gives the group velocity $v_g$ where

$$\frac{1}{v_g}=\frac{1}{c}\left(n+\omega\frac{dn}{dw}\right) \quad .$$

Near resonance $dn/dw<0$ and the group velocity can become negative; this is the classical case of anomalous dispersion. Garrett and McCumber[1] have shown theoretically that for $\omega\Gamma >> \Lambda^2$ a transform limited Gaussian pulse appears to propagate at $v_g$ even when $v_g$ is negative. This has recently been experimentally demonstrated by Chu and Wong.[2]

Secondly, we consider the case of strong coupling ($\Lambda^2 >> \omega\Gamma$) and finite spatial dispersion ($Dk^2>0$). The exciton and photon lose their separate identity and must be described as a polariton. The dispersion curve of the polariton consists of two branches with a positive group velocity in each branch, small near resonance. The appropriate boundary conditions determine the branch in which propagation predominantly occurs.[3] Time-of-flight measurements in GaAs,[4] CuCl,[5] and CdSe (Ref. 6) have shown large pulse delays corresponding to these low group velocities.

In the case of a layered structure such as the two-dimensional (2D) GaAs multi-quantum-well structure the mechanical exciton is spatially confined in a direction perpendicular to the layers, and the spatial dispersion for this direction is zero. For a picosecond pulse traveling normal to the layers we nevertheless observe velocity decreases at the exciton resonance which are comparable to those seen in bulk GaAs at similar optical power levels. Clearly spatial dispersion cannot be invoked to explain the behavior. Indeed, we would expect negative group velocities and hence a pulse advancement as in Ref. 2. However, we have found that hole burning can occur within the exciton line even at very low intensities ($\simeq10^{10}$ photons $cm^{-2}$ pulse). Hole burning has been shown to produce large pulse delays in many systems.[7,8] In this paper we show that it can account quantitatively for the observed delays. We also show that the measurements provide a useful technique for measuring damping and homogeneous linewidths under certain conditions in these systems.

The sample used in this experiment was grown by molecular beam epitaxy (MBE) on a GaAs substrate and consisted of 100 identical layers of GaAs, 51 Å thick, each separated by 200 Å of $Al_{0.25}Ga_{0.75}As$. A window was etched in the GaAs substrate to allow passage of the laser beam. A synchronously pumped modelocked dye laser gave pulses whose autocorrelation length $\tau$ could be varied from 9 to 30 psec. The corresponding spectral widths $\hbar\Gamma_L$ were 0.34 and 0.079 meV, respectively. The product $\tau\Gamma_L/2\pi$ was 0.77 to 0.55, close to the theoretical value of 0.48 for a $\mathrm{sech}^2(t)$ profile.[9] The incident energy density was varied from $10^{-7}$ to $10^{-9}$ J $cm^{-2}$ /pulse. The time of flight of the pulse through the GaAs layers was measured by the cross-correlation technique.[4] Measurements were made at temperatures from 2 to 60 K.

For a GaAs layer thickness of 51 Å the heavy exciton absorption is centered at 1.616 eV and has an inhomogeneous linewidth $\Gamma_x$ of 5 meV. Weisbuch *et al.*[10] attributed this width to fluctuations in the layer thickness. The delay of the peak of the pulse relative to the delay far-off resonance is plotted in Fig. 1 as a function of photon energy for two pulse widths. Note that $\Gamma_x >> \Gamma_L$. A plot of the absorption coefficient $\alpha$ is also included in the figure. Note that the maximum delay occurs 1 meV below the

 

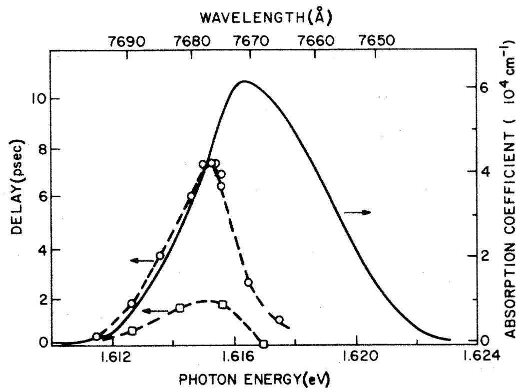


FIG. 1. Plot of the pulse delay and hole absorption coefficient $\alpha_H$ as a function of photon energy in the heavy exciton line for 51-Å GaAs layers. The incident power is $3\times10^{-8}$ J $cm^{-2}$/pulse. The open circles are for $\Gamma_L=0.079$ meV and the open squares for $\Gamma_L=0.34$ meV.

peak of the exciton line. For $\hbar\Gamma_L=0.079$ meV the maximum delay is 7.5 psec corresponding to a velocity of $6.7\times10^6$ cm $sec^{-1}$ through 0.5 $\mu$m of GaAs or about a factor of 2000 reduction in velocity. Spectrally broader pulses give a smaller delay.

When the pulse energy is decreased from its maximum value of $10^{-7}$ J $cm^{-2}$ /pulse the delay decreases only slightly at first. For the shortest pulse, $\tau=9$ psec, the maximum delay decreases from 2 psec at $10^{-7}$ J $cm^{-2}$ /pulse to 0.7 psec at $10^{-8}$ J $cm^{-2}$ /pulse. At $10^{-9}$ J $cm^{-2}$ /pulse the delay is less than 0.5 psec. For the longer pulse $\tau=29$ psec, $\hbar\Gamma_L=0.067$ meV, the delay does not change noticeably between $10^{-7}$ and $10^{-8}$ J $cm^{-2}$ /pulse below which intensity the signal was too low. Apart from a slight broadening at the lowest intensities (<10% of pulse length) near the peak of the delay, the pulse shape is unaltered.

The delay decreases monotonically with increasing temperature. At 16 K the delay is reduced by about a factor of 2 and at 60 K is too small to be observed. In this temperature range the shape of the exciton absorption remains unchanged except for a gradual shift to lower energy.

The dependence on temperature rules out the possibility of pulse-reshaping effects due to incomplete modelocking or pulse chirping. The observed delays are much larger than those expected in the linear regime. From Eq. (1) with $\Gamma=\Gamma_x$ an advance at line center of 0.25 psec is calculated; the maximum delay at the wings of the line would be 0.05 psec. Similarly, Loudon's energy velocity calculations[11] predict a delay of 0.3 psec at line center. Clearly these values are much too small to account for the data.

We now show that hole burning by the pulse in the exciton line provides a reasonable explanation of the data including the weak dependence on intensity. The depth and homogeneous width $\Gamma_{hom}$ of the hole were measured by a two-beam pump probe experiment. The details of these experiments are described elsewhere.[12] Table I summarizes the change in absorption coefficient $\alpha_H$ and the recovery rate $t_0^{-1}$ of the hole after the pump pulse for two different photon energies and two intensities. The recovery rates provide a lower limit to the homogeneous width of the hole $\Gamma_{hom}\simeq\tau_0^{-1}$. The effective hole width $\Gamma_H\approx\Gamma_L+\Gamma_{hom}$. $\Gamma_{hom}$ increases at higher photon energies and higher intensities. For $E_0>1.6176$ eV no hole could be observed at the available intensities, presumably because $\Gamma_{hom}$ becomes large. At $E=1.6153$ eV and $10^{-7}$ J $cm^{-2}$ /pulse $\alpha_H=-3.2\times10^4$ $cm^{-1}$ which is comparable with the absorption coefficient of $4.2\times10^4$ $cm^{-1}$ at this energy.

Selden[7] and Smith and Allen[8] have considered the delay of pulses propagating through saturable absorbers due to incoherent bleaching, but they did not consider the inhomogeneously broadened case where a hole can be burnt. The dependence on pulse width in the present case shows that the spectral width of the hole is indeed crucial. The pulse propagation is envisaged crudely as follows. As the pulse passes through the material the early part of the pulse burns a hole and is strongly attenuated. The hole, which can be viewed as a negative absorption sitting on top of a much broader resonance, gives rise to its own anomalous dispersion which can alter the group velocity of the remainder of the pulse. The change in group velocity and in the shape of the transmitted

TABLE I. Values of $\alpha_H$ and $\tau_0^{-1}$ for different incident powers and photon energies. $\alpha_H$ is in units of $cm^{-1}$ and $t_0$ in seconds.

| $I$ \ $E_L$ (eV) | | 1.6131 | 1.6153 | 1.6176 |
|---|---|---|---|---|
| $10^{-7}$ | $-\alpha_H$ | $10^4$ | $3.2\times10^4$ | $1.4\times10^4$ |
| J $cm^{-2}$/pulse | $t_0^{-1}$ | $5.6\times10^9$ | $4\times10^{10}$ | $>2.5\times10^{11}$ |
| $10^{-8}$ | $-\alpha_H$ | $<0.2\times10^4$ | $0.5\times10^4$ | $0.1\times10^4$ |
| J $cm^{-2}$/pulse | $t_0^{-1}$ | $4\times10^9$ | $1.8\times10^{10}$ | $>1\times10^{11}$ |

pulse will depend on the details of the hole width and depth and on the ratio of the laser to hole width. When the homogeneous width $\Gamma_{hom}$ of the hole is comparable to or greater than the laser width $\Gamma_L$ the pulse contains only frequencies near the peak of the hole. In this case we can put $\omega = \omega_0$ in Eq. (1) where $\omega_0$ is now the center frequency of the hole. Solving for the real part of the index of refraction gives the group velocity $v_g$ at the peak of the hole where

$$1/v_g = -\alpha_H/\Gamma_H \quad . \qquad (2)$$

Since $\alpha_H$ is negative, this gives rise to a pulse delay, and since $\Gamma_H \ll \Gamma_x$ the delay can far exceed the linear contribution. From Table I we can now calculate the delays expected from this model. At $E = 1.6153$ eV an incident intensity of $10^{-7}$ J cm$^{-2}$/pulse burns a 75% hole in the exciton line so that only a small part of the pulse is used to create the hole. Consequently, the propagation of most of the pulse will be determined by the presence of this hole. From Table I $\Gamma_{hom} \geq t_0^{-1} = 4 \times 10^{10}$ Hz. Since this represents a lower value for $\Gamma_{hom}$, the laser pulse width of 0.079 meV ($\Gamma_L = 1.2 \times 10^{11}$ Hz) used in the experiment is of comparable magnitude and the net width of the hole $\approx 1.6 \times 10^{11}$ Hz. This width is broader than the laser width and the assumptions underlying Eq. (2) are reasonably valid. Consequently, we expect the pulse to be transmitted largely unchanged in shape but delayed by $\approx 10$ psec as calculated for $\alpha_H = -3.2 \times 10^4$ cm$^{-1}$. This delay is very close to that observed. For $\Gamma_L = 0.34$ meV, on the other hand, a delay of 2.78 psec is calculated again in close agreement. As the intensity is decreased, $\alpha_H$ and $\Gamma_H$ both decrease so that the dependence of the delay on intensity is weak. This continues until $\Gamma_{hom} \ll \Gamma_L$ at which stage $\Gamma_H \doteq \Gamma_L$, independent of intensity. Our approximations at this point are no longer valid: nevertheless, the calculated and observed delays still agree to within a factor of 2.

At higher photon energies $t_0$ was too short to be measured accurately but we can use the theory to obtain $\Gamma_{hom}$. For $E = 1.617$ eV and $\Gamma_L = 0.079$ meV the delay is 1 psec; $\alpha_H = 1.6 \times 10^4$ cm$^{-1}$ so that $\Gamma_{hom} = 0.45$ meV. This corresponds to a minimum hole recovery time of $\approx 1.5$ psec.

At the lower-energy side of the line the recovery time of the hole is much longer than the pulse length, so that one of the conditions for self-induced transparency (SIT) is satisfied. We have used the theory of McCall and Hahn[13] to estimate the delay for a $2\pi$ pulse propagating through 0.5 $\mu$m of GaAs. For $E < 1.6153$ eV the estimated delays agree with the observed delay to within a factor of 2, but in the absence of further more concrete evidence such as pulse breakup the occurrence of SIT could not be verified.

The temperature dependence of the delay can be easily explained by the increase in $\Gamma_H$ as the temperature is raised. This can result from factors such as exciton dissociation or collisions due to increased mobility.

In conclusion, we have demonstrated that hole burning in the exciton line can lead to delays of propagating pulses comparable to the delays arising from linear effects. The weak dependence on intensity and the dependence on pulse width, temperature, and position in the line are all consistent with this model. In strongly absorbing media these nonlinear effects will be important even at low power densities and may easily be confused with the linear effects. Finally, we have shown that the pulse delay is a reasonable measure of the homogeneous linewidth with the condition that this width is greater than the laser linewidth, a condition that can be ascertained from a measure of $t_0$ in a simple pump-probe experiment.

I wish to thank M. D. Sturge and S. Chu for many helpful discussions and S. L. McCall for suggesting the relevance of nonlinearity in this system. I also wish to thank P. Hu and B. F. Levine for their stimulating interest and R. C. Dunne for excellent technical assistance. The samples were provided by A. C. Gossard, Bell Laboratories.

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