Surface Science 228 (1990) 69–73
North-Holland

HIGH RESOLUTION LASER SPECTROSCOPY OF RELAXATION AND THE EXCITATION LINESHAPE OF EXCITONS IN GaAs QUANTUM WELL STRUCTURES

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Received 2 June 1989; accepted for publication 18 August 1989

A new class of measurements on GaAs quantum well structures based on frequency domain nonlinear laser spectroscopy is described. Room temperature measurements of the excitation relaxation show an interference effect in the lineshape which is interpreted as a shift in exciton frequency. Low temperature measurements on the localized exciton provide an excitation lineshape which eliminates the effects of inhomogeneous broadening and shows the presence of spectral diffusion.

The room temperature nonlinear optical response near the band edge in GaAs quantum wells has resulted in considerable interest in these materials because of their potential importance to optoelectronic devices [1]. Studies of these materials have also resulted in new understanding of the fundamental physics of excitons [2–4]. In this paper, we describe a new class of measurements based on high resolution frequency domain nonlinear laser spectroscopy which are distinguished from earlier measurements because of the ability to report on excitation lineshapes as well as the ability to provide greater sensitivity in measurements of excitation relaxation. The measurements of the excitation lineshape are significant because this spectroscopy eliminates the contribution of inhomogeneous broadening which characterizes low temperature absorption measurements.

The basic experimental configuration for these experiments is based on backward four wave mixing (FWM) [5]. In these experiments, two optical beams intersect each other at a small angle. One beam is designated the probe beam with field amplitude $\boldsymbol{E}_{\mathrm{p}}(\omega_{\mathrm{p}}, \boldsymbol{k}_{\mathrm{p}})$ while the remaining beam is the forward pump beam with field amplitude $\boldsymbol{E}_{\mathrm{f}}(\omega_{\mathrm{f}}, \boldsymbol{k}_{\mathrm{f}})$. The beams are nearly normal to the growth direction of the quantum well and produce an interference pattern which results in a spatial and temporal modulation of excitation proportional to $\boldsymbol{E}_{\mathrm{f}}(\omega_{\mathrm{f}}, \boldsymbol{k}_{\mathrm{f}}) \cdot \boldsymbol{E}_{\mathrm{p}}^{*}(\omega_{\mathrm{p}}, \boldsymbol{k}_{\mathrm{p}})$. A coherent signal, $\boldsymbol{E}_{\mathrm{s}}(\omega_{\mathrm{s}}, \boldsymbol{k}_{\mathrm{s}})$, is produced by the scattering of a back beam with field amplitude $\boldsymbol{E}_{\mathrm{b}}(\omega_{\mathrm{b}}, \boldsymbol{k}_{\mathrm{b}})$. Spectroscopic information is obtained by varying any one frequency with respect to the remaining two frequencies. In particular, the FWMp response is obtained by varying ω_{p} holding $\omega_{\mathrm{f}} = \omega_{\mathrm{b}}$ while the FWMb response is obtained by varying ω_{b}, holding $\omega_{\mathrm{p}} - \omega_{\mathrm{f}} = \text{constant}$. The FWMp response provides a lineshape related to the excitation decay rate and contains contributions such as inelastic scattering and carrier or excitonic recombination in addition to the contribution of nonradiating states. The FWMb response provides a measure of the excitation lineshape without contributions from inhomogeneous broadening [5,6].

At room temperature, the exciton absorption lineshape is dominated by homogeneous broadening due to the rapid LO phonon ionization rate. Hence, a study of the FWMb response provides little additional information. However, the nonlinear response arises because the electron–hole plasma generated from the ionization of the exciton results in band filling and exchange effects which lead to a modification of the optical proper-

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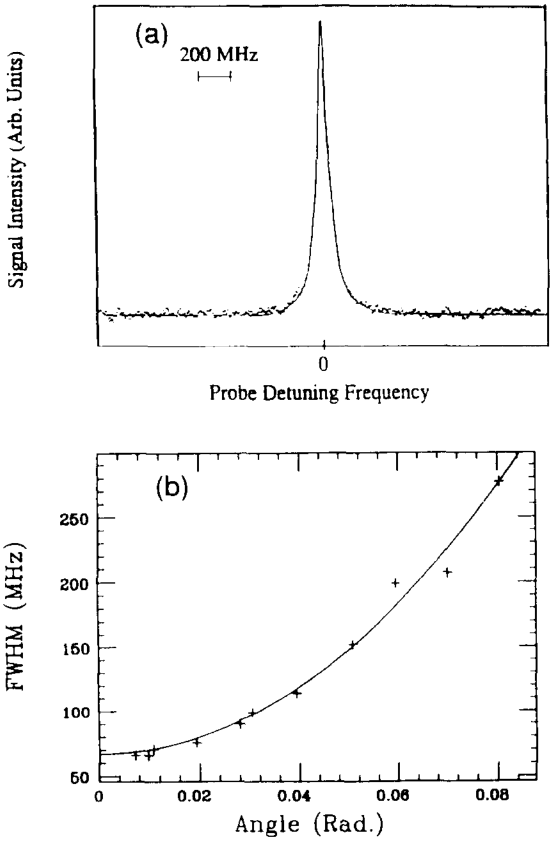

Fig. 1. (a) Room temperature FWMp lineshape. (b) Angle dependence of the FWMp line width.

ties of the exciton including the oscillator strength, dephasing rate, and resonance frequency. Hence, a grating produced by the interference of the forward pump and probe has a decay rate determined by the e–h recombination time and the time it takes carriers to spatially diffuse across the grating. Fig. 1a shows the FWMp response at room temperature along with a fit of a Lorentzian. The line width is given by $\gamma_{e-h} + D\,|\Delta \boldsymbol{k}|^2$ where γ_{e-h} is the e–h recombination rate, D is the ambipolar diffusion coefficient, and $|\Delta \boldsymbol{k}| = |\boldsymbol{k}_f - \boldsymbol{k}_p|$. Fig. 1b shows the angle dependence of the FWMp line width as a function of angle and the solid line is a fit of the above line width expression. From the shape of the curve, we can obtain the ambipolar diffusion coefficient (18 cm^2/s) and from the y-intercept, we can obtain the e–h recombination rate (5 ns).

Using the method of correlated optical fields [7] to eliminate interlaser jitter contributions to the lineshape, we examined the FWMp response near $\omega_p - \omega_f = 0$. Fig. 2 shows the lineshape. There are two important observations which can be drawn from fig. 2a. First, there exists a strong (dominant) contribution to the room temperature nonlinear response which is slow (10 μs). Secondly, the lineshape is characterized by a shape which is typical of interference effects [#1]. We believe this lineshape is due to a shift in the exciton resonance. Solutions to the optical Bloch-type equations which have been phenomenologically modified to account for band filling and exchange effects show that this interference profile results when the Bloch equations are further modified to account for a shift in the exciton resonance [9]. The curve in fig. 2b is the calculated FWMp response, showing the interference lineshape. It is important to stress that the solutions of the modified Bloch equations only show interference effects when the exciton resonance is allowed to shift due to the presence of the e–h plasma or some other laser induced mechanism. We also note that the shape of the FWMp response is dependent on the wavelength of the back pump. Fig. 2c shows the FWMp lineshape measured when the back pump is tuned red of the peak of the heavy hold exciton. (The forward pump and probe beams remained tuned on the peak of the heavy hold exciton.) Fig. 2d shows the lineshape calculated from the modified Bloch equations when the back pump is shifted red of the exciton resonance.

At low temperatures, the excitons are stable against phonon ionization. Hence, studies of relaxation provide information on the exciton dynamics. In addition, because the exciton is now stable, data obtained from the FWMb response provide information on the excitation lineshape. At 5 K, the absorption lineshape is inhomogeneously broadened due to fluctuations in the well thickness. The HH1 absorption line width in this sample is 3.7 meV. The sample was further characterized by performing luminescence measure-

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[#1] Interference effects are common in nonlinear laser spectroscopy as discussed in ref. [8]. However, the origin of the interference effects in this paper is new and distinct.

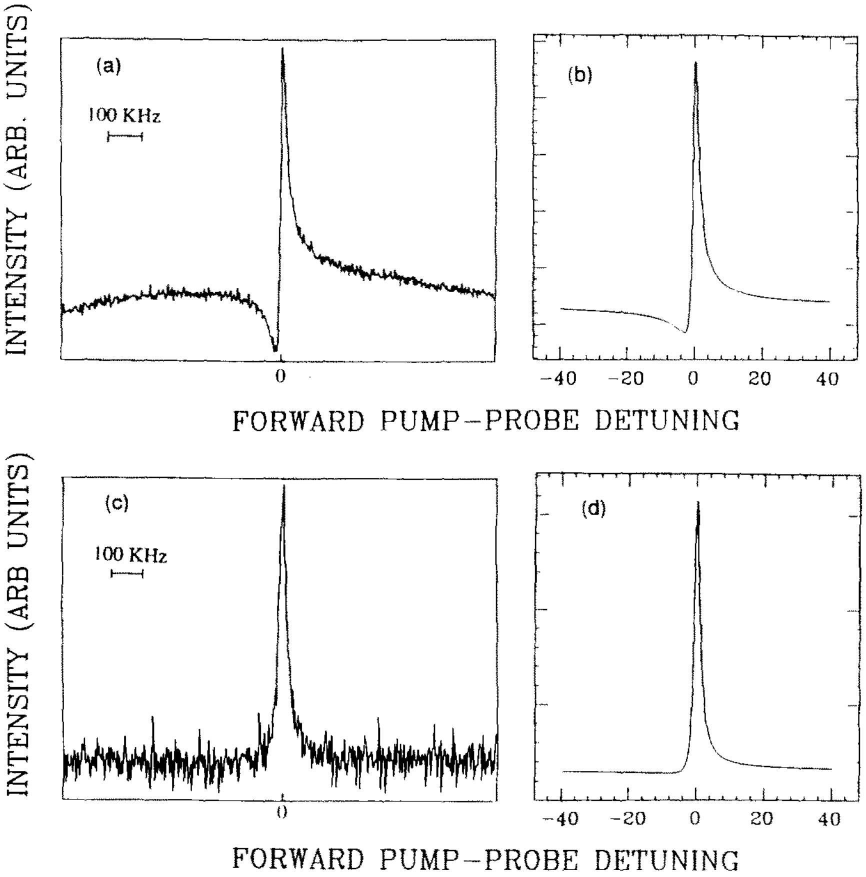

Fig. 2. The FWMp line shape using the technique of correlated fields. (a) Interference profile when the back beam wavelength is on the exciton peak. (c) The lineshape when the back beam is tuned red of the exciton peak. (b, d) Calculated lineshapes using modified Bloch equations.

ments as well as obtaining a degenerate four wave mixing (DFWM) spectrum. (A DFWM spectrum is obtained by using one laser for all beams in the nonlinear interaction and observing the change in intensity of the signal as the frequency of the laser is tuned across the exciton resonance.) The peak of the luminescence from the excitons is shifted 2.8 meV to the red of the HH1 absorption maximum. The red shifted luminescence is interpreted as being due to the recombination of excitons which have become localized due to fluctuations in well thickness. The DFWM measurements show a peak which coincides exactly with the luminescence peak, suggesting the nonlinear response is also due to localized excitons.

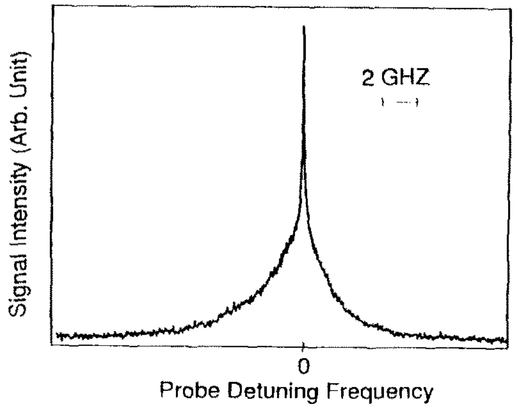

Fig. 3. The FWMp lineshape at 5 K showing the presence of multiple relaxation mechanisms.

In the first set of measurements, we examined the FWMp response to determine the excitation relaxation dynamics. In the simple picture of the exciton undergoing a single relaxation due to radiative decay, the lineshape would be a single Lorentzian. Fig. 3 shows the FWMp line width obtained at 5 K. The curve shows clear evidence of multiple decay processes rather than a single decay. In particular, three decay channels are observed in fig. 3. The fastest and slowest channels having relaxation times of 100 ps and 15 ns are clearly observed in this figure. However a third component to the lineshape corresponding to a relaxation time of 1.5 ns is observable upon expansion of the center portion of the figure. The 1.5 ns feature corresponds to the expected radiative decay time for a well of thickness of 98 Å [10]. The origin of the 100 ps feature is most likely due to decay of the excitation by spectral diffusion via phonon assisted tunneling. Similar behavior has been reported in transient pump–probe measurements [4]. Further evidence for this is given below. The origin of the 15 ns decay rate is due possibly to the presence of electric fields in the material which are known to extend the life time of the exciton.

FWMb experiments were then conducted to determine the lineshape of the excitation. Note that the energies at which the following FWMb lineshapes were recorded are referenced with respect to the position of the peak of the absorption. Fig. 4a shows the FWMb lineshape obtained 3.45 meV to the low energy side of the absorption maximum. The lineshape is slightly asymmetric on the high energy side. This asymmetry becomes much clearer in the FWMb spectra recorded at the absorption line center as seen in fig. 4b. The presence of asymmetry in the FWMb response is highly significant, since it can be shown [6] that if we are measuring just the homogeneous lineshape, the FWMb response should be symmetric about $\omega_b - \omega_f = 0$, even if the homogeneous lineshape is asymmetric. However, such asymmetric lineshapes can be observed in the FWMb response in the presence of spectral diffusion. Hence we believe these measurements show directly that excitons are spectrally diffusing, as suggested by earlier work [4]. Fig. 4c shows a comparison of the measured temperature dependence of the line width in fig. 4a to a theory based on phonon assisted tunnelling. The curve varies as $e^{\beta T^{1.6}}$ and is in general agreement with theoretical expectations [11].

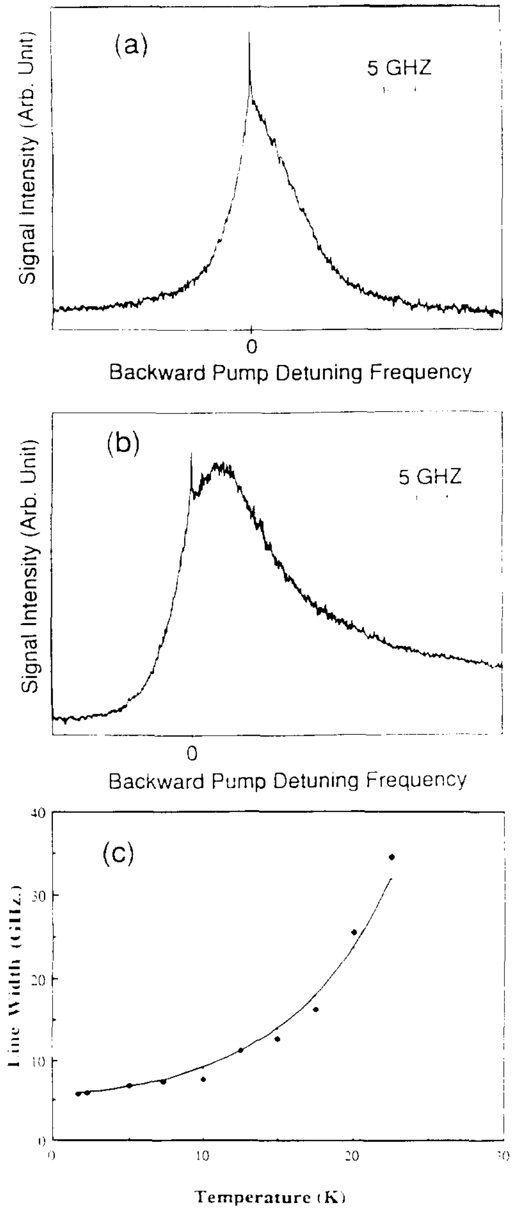

Fig. 4. The FWMb line shape at 5 K. (a) The lineshape red of the absorption maximum. (b) The lineshape measured at the peak of the exciton absorption. (c) The temperature dependence of the line width in (a).

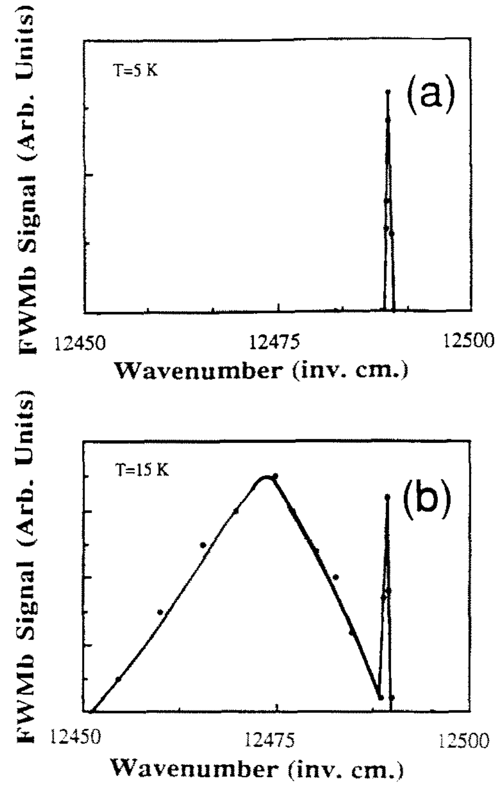

Fig. 5. FWMb response shown for 2 temperatures. (a) The response at 5 K. (b) The response at 15 K illustrating the spectral diffusion of the excitation to lower energies.

Further evidence for spectral diffusion is shown in fig. 5. Fig. 5a is an extended scan of the data shown in fig. 4b at 5 K. Fig. 5b shows the same data taken at 15 K illustrating the spectral diffusion of excitons to lower energies, the peak of the response occurring at the same energy as the peak of the luminescence. As in the case of the luminescence peak, the wide resonance in the FWMb spectra shown in fig. 5b represents the energy distribution of the localized excitons. Theoretical results of the FWMb line shape obtained from optical Bloch-type equations, modified to include spectral diffusion, show similar behavior. It is interesting to note that while we obtain clear evidence for energy diffusion, there is no evidence in the data to suggest that the excitons are spatially diffusing.

In summary, we have used high resolution frequency domain nonlinear spectroscopy to study relaxation and excitation lineshapes in GaAs multiple quantum wells. At room temperature, we have measured the ambipolar diffusion coefficient and the free carrier recombination time. In addition, we have observed an interference effect in the FWMp lineshape which we believe is due to a shift in the exciton resonance. At low temperature, we have identified contributions to the relaxation of the exciton population due to spectral diffusion and exciton recombination. Using FWMb spectroscopy, we have obtained measurements of the lineshape of the excitation. The observation of asymmetric FWMb lineshapes and the measured temperature dependence of the line width provide evidence that the excitons are spectrally diffusing via phonon assisted tunnelling.

This work is supported by the AFOSR, US ARO and the US ARO URI.

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