Optical Two-Dimensional Fourier Transform Spectroscopy of Excitons in Semiconductor Nanostructures

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The coherent opticalresponse of direct gap semiconductors is complex due to the interplay between many-body effects and disorder. At low temperature, the optical response close to the fundamental band gap is dominated by excitonic resonances. Extensive studies using traditional coherent spectroscopic techniques, such as transient four-wave-mixing (TFWM), have shown that many-body phenomena are dominant [1].Recently, optical two-dimensional Fourier transform spectroscopy (2DFTS) has been used to distinguish amongst the various many-body contributions in GaAs quantum well structures [2,3], which could not easily be separated using TFWM. Comparison between experiment and theory clearly shows that many-body correlation terms beyond Hartree-Fock play an essential role. Many-body interactions compete with disorder, which tends to localize excitons and thus reduce their interactions. In the optical response, disorder is manifest as inhomogeneous broadening of the exciton resonance. 2DFTS is predicted to be able to simultaneously measure the homogeneous and inhomogeneous linewidths of the exciton resonances [4].

Using a new apparatus, we have been able to reduce the optical excitation density so that the effects of disorder become apparent in the experimental results. The diagonal elongation of the exciton resonances due to disorder is clearly observed. Variation is expected due to a transition from localized states on the low energy side of the inhomogeneous line to delocalized states above it. We observe a peak shifted in the emission dimension by about 2 meV, which we attribute to biexcitons. The biexciton line also shows diagonal elongation, suggesting that biexcitons primarily form from two excitons on the same localization site.The elongation of the biexciton line is not exactly along the 45° diagonal, which shows that biexciton binding energy is not constant, but rather is greater for low energy states.

Two dimensional spectra are actually a slice of a three-dimensional spectrum, where the dimensions correspond to the delay between the first and second pulse, t, the delay between the second and third pulses, T, and the emission time, t. The standard 2D

spectra are taken by holding *T*constant and Fourier transforming the signal obtained as a function of *t* and *t*. By holding t constant and Fourier transforming the signal as a function of *T* and *t*, it is possible to isolate non-radiative coherences including Raman and two-quantum coherences.These spectra allow the decoherence times of the non-radiative coherences to by determined directly, which is not possible using one-dimensional techniques.

REFERENCE

[1] For a recent review, see S.T. Cundiff, Opt. Express **16,** 4639-4664 (2008).

[2] X. Q. Li, T. Zhang, C. N. Borca, and S. T. Cundiff, Phys. Rev. Lett. **96**, 057406 (2006).

[3] T. Zhang, I. Kuznetsova, T. Meier, X. Li, R. Mirin, P. Thomas, and S.T Cundiff, Proc. Nat. Acad. Sci. **104**, 14,227-14,232 (2007).

[4] I.Kuznetsova, T. Meier, S. T. Cundiff, and P. Thomas, Phys. Rev. B **76**, 153301 (2007).