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Femtosecond time-resolved four-wave mixing from biexcitons in GaAs quantum wells: Dominance of the interaction-induced signal

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Time-resolved (TR) four-wave mixing (FWM) experiments are performed on a high-quality GaAs quantum-well sample where biexcitons make the dominant contribution under certain experimental conditions. TR-FWM peaks well after both beams passed through, *regardless* of relative contributions of biexcitons to FWM. Our experiments show that the diffraction of the interaction-induced field dominates FWM from biexcitons, as well as from excitons.

In the past few years, the importance of the coherent exciton-exciton interaction in nonlinear optics of semiconductors has been recognized by the existence of strong time-integrated (TI) four-wave mixing (FWM) signal in negative time delays in high-quality GaAs quantum wells (QW's).[1–3] Recently, time-resolved (TR) FWM (Ref. 4) showed that the interaction-induced field ($E_{\rm II}$) completely dominates FWM from GaAs QW's at low temperatures: the signal continued to increase after both beams passed through and peaked at $t \approx T_2$ measured from the arrival of the second beam (Fig. 1). The predictions and results of Refs. 1–4 are in stark contrast with the predictions of the noninteracting two-level model[5] where the diffraction of the incident field (E_0) determines FWM. In this model, the negative time delay signal does not exist and the peak of TR-FWM would always occur at $t \approx t_p$ (t_p is the pulse width), regardless of T_2. However, they are in excellent agreement with microscopic theory of FWM.[6,7]

In samples used in Refs. 1–4, the biexcitonic effect in FWM was weak or negligible, confirmed by the intensity dependence of cw luminescence, TI-FWM, or spectrally resolved (SR) FWM.[8,9] However, in some samples, there exists strong biexcitonic contribution to FWM.[10–14] Often, this is accompanied by the existence of the negative time delay signal whose time constant is equal to or larger than that of the positive time delay signal.[10,11] This is explained by the creation of biexcitons by beam 2 (which precedes beam 1 for $T<0$), and the subsequent diffraction of beam 1 into the direction of $2k_2-k_1$. This way, the existence of the strong negative time delay signal in FWM can be explained. Another sign is the appearance of an additional peak or shoulder at the lower-energy side[11,12,16] in SR-FWM. Also, the biexcitonic binding energy has been estimated by attributing the observed quantum beats to the exciton-biexciton system.[13] There exists a wide range of values for the biexcitonic binding energy, from 0.35 to 2.7 meV, depending on the width of QW's or some other (unknown) parameters.[10–14]

It is not clear why the relative biexcitonic contribution or the binding energy are strongly sample dependent. While we do not try to resolve this question in this paper, a fundamental issue remains unresolved. Most results in the biexcitonic contribution to FWM are analyzed using the *noninteracting* model of Ref. 5 where the *diffraction of $E_{\rm II}$ is neglected.* This is understandable because in samples with strong biexcitonic contribution to FWM, the existence of a negative time delay signal itself does not require the diffraction of $E_{\rm II}$. However, considering the fact that biexcitons are created by exciton-exciton interaction, and in light of the now well-established importance of exciton-exciton interaction in FWM, this simple approach ignoring the diffraction of $E_{\rm II}$ *warrants further experimental investigations.* To resolve this issue directly and conclusively, time evolution of FWM signals originating from biexcitons should be studied.

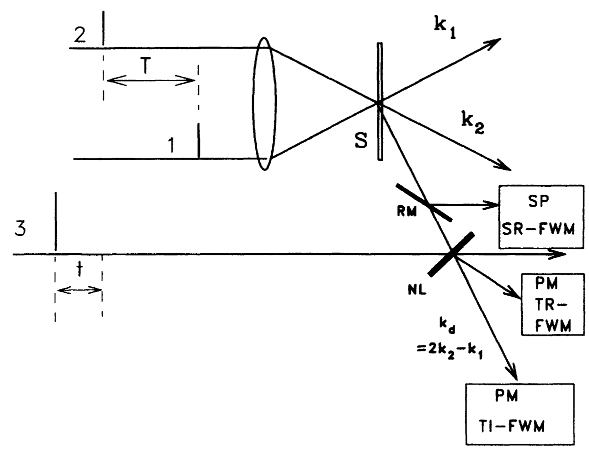

FIG. 1. Schematics for TI-, TR-, and SR-FWM experiments. In TR-FWM, the diffracted signal in the direction of k_d is up-converted in a nonlinear crystal (NL) with a third beam. The time delay thus introduced (t) acts as a real time, so that time evolution of FWM signal at a fixed time delay (T) can be probed. S, SP, RM, and PM denote sample, spectrometer, removable mirror, and photomultiplier tube, respectively.

In this paper, we have performed femtosecond (100–200 fs) TI-, SR-, and TR-FWM experiments (Fig. 1) on high-quality GaAs QW's. This sample has an absorption width at the heavy-hole (HH) exciton which is less than 1 meV and has well widths of 170 Å (15 wells, 0.9 meV). Because of relatively small total thickness, pulse distortion in this sample is minimal.[15] When two cross-linearly polarized beams are used at resonance, biexcitonic contribution dominates FWM in this sample, even at modest excitation densities ($<10^{10}$ cm^{-2}). This is consistent with the recent results[10] where a large enhancement of biexcitonic effect in resonant excitation conditions ("cold excitons") relative to the off-resonance conditions was reported. Furthermore, in our sample, FWM signals in the cross-linearly polarized geometry are at least as strong as or even stronger than those in the collinearly polarized geometry, which is consistent with the strong biexcitonic contribution in the cross-linearly polarized geometry. The same geometry was used in recent works[10,11] where strong biexcitonic effects in FWM were reported. Our TR-FWM results show that the peak of TR-FWM signal occurs well *after both beams passed through*, regardless of whether excitons (in the collinearly polarized geometry) or biexcitons (in the cross-linearly polarized geometry) dominate FWM. Our results unambiguously demonstrate that diffraction of the induced field dominates FWM from biexcitons as well as from excitons.[4] From these results, we conclude that the commonly used noninteracting model description of biexcitonic contribution is completely inadequate and a model including the diffraction of the interaction-induced field should be used.

In Fig. 2(a), TI-FWM from sample *B* in the cross-linearly polarized geometry are plotted at a moderate exciton density of $\approx 10^{10}$ cm^{-2} and 10 K. The laser is tuned 4 meV below the HH exciton (top), or at resonance with HH exciton (middle). At the bottom, the cross-correlated pulse shape after transmission is shown, with little distortion. It is clear that the time constant for $T<0$ is comparable (at 0-meV detuning) or larger (at -4-meV detuning) than those for $T>0$. This is consistent with the results of Refs. 10 and 11 and can be a sign of strong biexcitonic contribution. Figure 2(b) shows SR-FWM spectra at, from top to bottom, $T=-0.5$, 0, and 1 ps, and the laser spectrum is denoted by broken lines. The biexcitonic peak ≈ 1 meV below the excitonic peak dominates FWM in the cross-linearly polarized geometry (solid lines) at all time delays, while FWM in the collinearly polarized geometry (broken lines) is dominated by the excitonic contributions. Light-hole excitonic contribution is also visible, roughly 5 meV above the HH. These results are consistent with Refs. 10 and 11 where cross-linearly polarized beams were used to observe strong biexcitonic contribution to FWM. Further TI- and SR-FWM studies on this sample using circularly as well as linearly polarized beams reveal that this peak indeed corresponds to biexcitonic contribution to FWM.[16] Therefore, performing TR-FWM on this sample in the *cross-linearly polarized geometry* would definitely show whether the incident or the induced field dominates FWM originating from biexcitons.

In Fig. 3, the results of TR-FWM on sample *B* under comparable conditions with Figs. 2(a) and 2(b) are shown at various time delays. Since cross-linearly polarized beams are used, FWM is dominated by biexcitons. The peaks occur

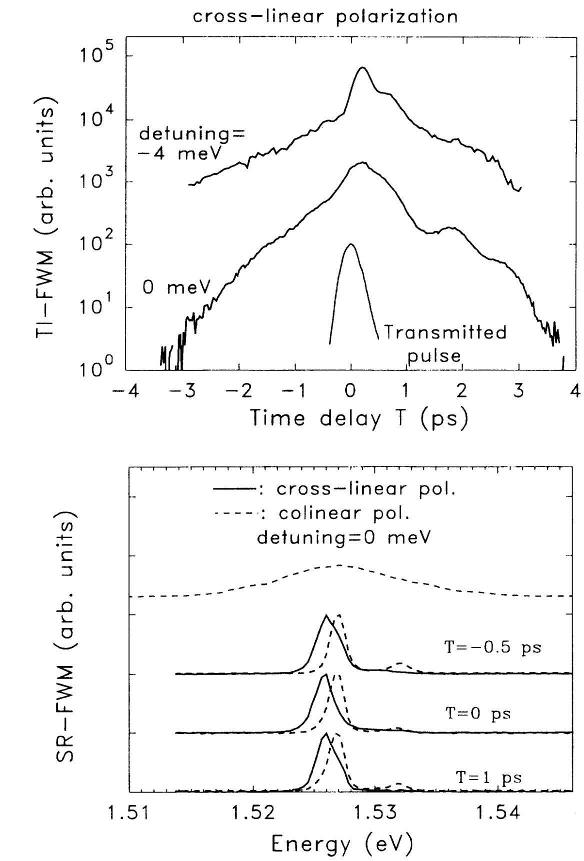

FIG. 2. (a) TI-FWM from sample *B* at 10 K in the cross-linearly polarized geometry at detunings of -4 meV (top) and 0 meV (middle) from the HH resonance. At the bottom, the transmitted pulse shape determined by the cross correlation in a nonlinear crystal is shown. (b) SR-FWM for the cross and the collinear polarization geometries at $T=-0.5$, 0, and 1 ps. The density is estimated to be $\approx 10^{10}$ cm^{-2}. The laser spectrum is denoted as broken lines at the top.

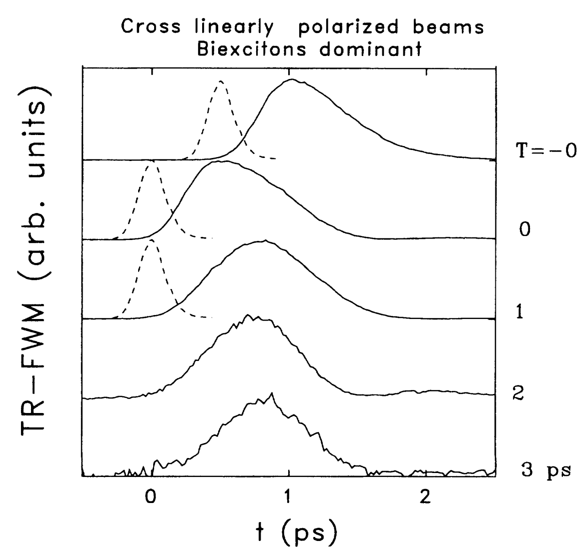

FIG. 3. TR-FWM from sample *B* under nearly identical conditions as Fig. 2(b). Time delays are, from top to bottom, -0.5, 0, 1, 2, and 3 ps.

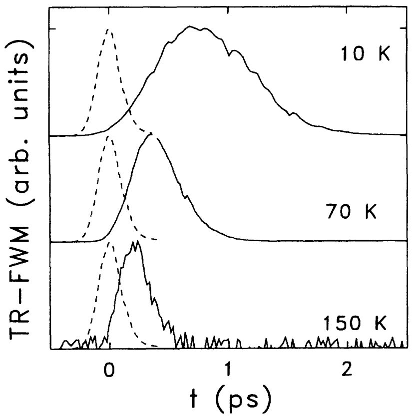

FIG. 4. Temperature dependence of TR-FWM from sample B at $T=1$ ps. The experimental conditions are nearly identical with those of Fig. 3. The temperatures are, from top to bottom, 10, 70, and 150 K.

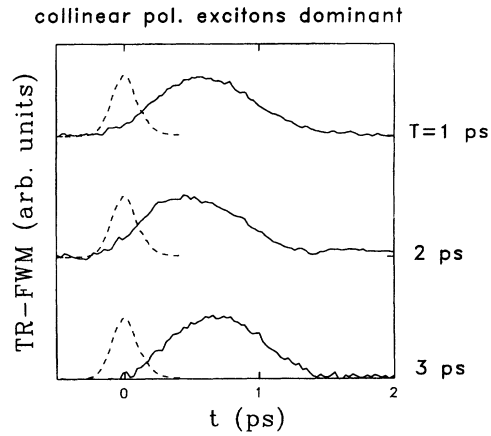

FIG. 5. TR-FWM at 10 K in the collinearly polarized geometry at $T=1$, 2, and 3 ps. The density is estimated to be $\approx 10^{10}$ cm^{-2}.

well away from the passage of the second beam, both for positive and negative time delays, whose positions and transmitted shapes are denoted by broken lines. From this, we can immediately conclude that the diffraction of E_{II} completely dominates FWM from biexcitons, as well as from excitons.[4]

Our conclusion is further supported by the temperature dependence of TR-FWM shown in Fig. 4. Since TR-FWM resulting from the diffraction of E_{II} would peak at $t\approx T_2$, while that from the diffraction of E_0 would peak at $t\approx t_p$, performing temperature dependence of TR-FWM is a very powerful way of determining the relative importance of diffractions of E_0 and E_{II} in FWM.[4] The peak positions in Fig. 4 occur close to $t=0$ as temperature is increased (and therefore as T_2 is decreased), in agreement with the dominance of diffraction of E_{II}. At higher temperatures, biexcitons would dissociate and excitons, not biexcitons, would make the dominant contribution.[10–13] Therefore, it is expected that TR-FWM at 70 and 150 K are mostly from excitons, whereas at 10 K, biexcitons dominate. At 150 K, the experiment is almost time resolution limited. Therefore, the peak position at 150 K ($t\approx 0.14$ ps) sets an upper limit to the expected peak positions if the diffraction of E_0 were to make the dominant contribution, as the noninteracting model[5] would predict.

In Fig. 5, we show TR-FWM at $T=1$, 2, and 3 ps in the collinearly polarized geometry. In this geometry, excitons rather than biexcitons make the dominant contribution, as can be readily seen in Fig. 2(b). The peaks of TR-FWM again occur completely delayed from the arrival of the second pulse at $t=0$, denoted by broken lines. This is entirely consistent with the results of Ref. 4 obtained from a different sample which does not show strong biexcitonic contribution even in the cross-linearly polarized geometry.

In the microscopic theory of FWM, any electric field inside a semiconductor can be the source of the diffraction.[1–4,6,7] Therefore, the interaction-induced fields created by the Coulomb interactions among and between coherent excitons and biexcitons can be diffracted as well as the incident laser field. While it is clear that further theoretical and experimental studies are needed, our experiments demonstrate that including the diffraction of the induced field is essential for proper description of FWM from biexcitons. This is not so surprising considering the fact that the dominance of E_{II} in the excitonic contribution to FWM is now well known. In fact, it would have been rather surprising if the noninteracting description of FWM from biexcitons were the correct picture, while FWM from excitons is completely dominated by the diffraction of the interaction induced field E_{II}.[1–4] It is therefore desirable that a microscopic model including coherent interactions among excitons and biexcitons be used for adequate description of FWM originating from biexcitons. It is expected that such calculations would be difficult and time consuming. On the other hand, as recently proposed,[17] a considerable simplification can be achieved when an interacting four-level model is used, which may retain most of the essential physics.

In conclusion, we have performed TR-FWM experiments on a high-quality GaAs quantum-well sample where biexcitons dominate the FWM in the cross-linearly polarized geometry. Interaction-induced fields dominate FWM regardless of the relative contributions, from negligible to dominant, of biexcitons. This conclusion was drawn by realizing that the peak positions of TR-FWM always occur well beyond the expected peak positions of $t\approx t_p\approx 0.1$–0.2 ps if the diffraction of E_0 were to dominate FWM. Our results show that to describe FWM from biexcitons, the coherent interactions among excitons or biexcitons should be included.

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[1] K. Leo, M. Wegener, J. Shah, D. S. Chemla, E. O. Göbel, T. C. Damen, S. Schmitt-Rink, and W. Schafer, Phys. Rev. Lett. **65**, 1240 (1990).

[2] M. Wegener, D. S. Chemla, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. A **42**, 5675 (1990).

[3] K. Leo, E. O. Göbel, T. C. Damen, J. Shah, S. Schmitt-Rink, W. Schäfer, J. E. Müller, K. Köhler, and P. Ganser, Phys. Rev. B **44**, 5726 (1991).

[4] D. S. Kim, J. Shah, T. C. Damen, W. Schäfer, F. Jahnke, S. Schmitt-Rink, and K. Köhler, Phys. Rev. Lett. **69**, 2725 (1992).

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

[6] C. Stafford, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. B **41**, 10 000 (1990).

[7] W. Schäfer, F. Jahnke, and S. Schmitt-Rink, Phys. Rev. B **47**, 1217 (1993).

[8] K. Leo, T. C. Damen, J. Shah, and K. Köhler, Phys. Rev. B **42**, 11 359 (1990).

[9] D. S. Kim, J. Shah, T. C. Damen, W. Schäfer, and K. Köhler (unpublished).

[10] D. Lovering, R. T. Phillips, and G. J. Denton, Phys. Rev. Lett. **68**, 1880 (1992).

[11] B. F. Feuerbacher, J. Kuhl, and K. Ploog, Phys. Rev. B **42**, 2429 (1991).

[12] R. Raj, I. Abram, and J. A. Levenson, Solid State Commun. **81**, 51 (1992).

[13] S. Bar-Ad and I. Bar-Joseph, Phys. Rev. Lett. **68**, 349 (1992).

[14] M. Kuwata-Gonokami, in *International Quantum Electronics Conference (IQEC), Anaheim, California, 1994*, Technical Digest Series Vol. 9 (Optical Society of America, Washington, D.C., 1994); G. O. Smith, E. J. Mayer, and J. Kuhl, *ibid.*

[15] D. S. Kim, J. Shah, D. A. B. Miller, T. C. Damen, and L. N. Pfeiffer, Phys. Rev. B **48**, 17 902 (1993).

[16] H. Wang, J. Shah, T. C. Damen, and L. N. Pfeiffer, Solid State Commun. (in press).

[17] K. Bott, O. Heller, D. Bennhardt, S. T. Cundiff, P. Thomas, E. J. Mayer, G. O. Smith, R. Eccleston, J. Kuhl, and K. Ploog, Phys. Rev. B **48**, 17 418 (1993).