# Femtosecond Four-Wave Mixing Experiments on GaAs Quantum Wells Using Two Independently Tunable Lasers

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Femtosecond two beam and three beam ($\omega_1, \omega_1; \omega_2$) four-wave mixing (FWM) experiments on GaAs quantum wells have been performed using two partially synchronized, independently tunable lasers with external jitter compensation. Heavy and light hole beatings are observed with these two mutually incoherent lasers. FWM signals are observed when $\omega_2$ is completely below the exciton energies, with no spectral overlap with the absorption profile. These off-resonant signals are stronger than the interband continuum signals for equivalent detunings. [S0031-9007(98)06123-7]

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In the past few years, femtosecond nonlinear spectroscopy of semiconductors and semiconductor quantum structures have provided new insights on incoherent and coherent dynamics. This was possible mainly through the proliferation of femtosecond sources such as the self-modelocked Ti:Sapphire lasers, whose tuning range encompasses band gaps of GaAs, AlGaAs, InGaAs, CdTe, and other important semiconductors. One of the most widely used tools has been the two beam and three beam four wave mixing (FWM) [1]. There exists a wealth of information provided by this technique, which includes the relaxation of excitons, free carriers, and phonons, and interactions among them [2,3]. The wide tunability and the ease of second harmonic generation using femtosecond Ti:Sapphire lasers further extended the usage of FWM into wide band gap materials such as ZnSe [4] and GaN [5].

Most of the femtosecond wave mixing experiments have essentially been "one color" in that one performs, say, degenerate wave mixing experiments and tune the photon energy. In picosecond and subpicosecond high power experiments, synchronously pumped two dye lasers or Raman shifted lasers were used to investigate phonon dynamics in semiconductors [6]. In the femtosecond regime, Cundiff *et al.* performed partially nondegenerate FWM experiments by taking advantage of the broad bandwidth of sub-100 fs laser [7]. To perform femtosecond two color wave mixing experiments with a wide range of tunability, one needs to synchronize two femtosecond lasers with timing jitters between them as short as the pulse width. Clever techniques to generate two color femtosecond beams from one laser [8] or to synchronize two lasers with cavity length detuning exist [9], but with limited tunability. Commercially available systems deliver easy synchronization over the entire tuning range of 700–1000 nm between two femtosecond Ti:Sapphire lasers. However, the timing jitter is in the range of 5–10 ps.

In this Letter, we have performed the first two color femtosecond FWM experiments using two independently tunable, partially synchronized lasers. This was made possible by devising an easy way to externally compensate the inevitable jitters between the two lasers. We improved the effective time resolution from >5 ps to 150 fs, limited only by the pulse widths of the two lasers. Using this technique, we have performed femtosecond two beam and three beam FWM experiments on GaAs quantum wells. Our most interesting results are as follows: (1) Coherent phenomena such as the heavy-hole–light-hole (HH-LH) beating can be observed with two separate lasers, as long as the timing jitter can be improved. This holds true for both the two beam and three beam FWM geometries. (2) When the two beams from the first laser at $\omega_1$ form a population grating near zero delay, the second beam at $\omega_2$ can be diffracted over a very broad spectral range, including completely below the band gap. Furthermore, the signal at a given detuning below the band gap is larger than that for the equivalent detuning above. (3) When $\omega_2$ is completely above or below the excitonic resonance, the spectrum of the diffracted beam is mostly determined by the laser spectrum.

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In Fig. 1, we schematically describe the experimental setup. It is a standard degenerate two and three beam four-wave mixing configuration, except for the fact that in the two beam FWM, the second beam is delivered by another independently tunable laser. In the three beam FWM, the third beam is generated by the second laser. Since the timing jitter between these two commercially synchronized lasers is about 5–10 ps, performing two color femtosecond spectroscopy requires a drastic improvement. To this end, we generate sum frequency with these two lasers, and feed the sum frequency signal together with the FWM signal to a homemade "sample-and-hold" electronic switch, so that the FWM data are taken only when the two pulses are at a certain specific delay. Our technique is similar to that of Crooker *et al.* [10] who utilized acousto-optic modulator together with sum frequency generation. Our scheme is simpler and one major advantage is that lock-in amplifiers can be used despite the jitters because of the nature of "sample and holding." Upon using this technique, the cross correlation signal between the lasers, which was about 8 ps wide because of the jitters, was reduced to about 220 fs full width at half maxima (FWHM), corresponding to a 150 fs pulse width. The rms value for the jitter between the two laser pulses after the sample-and-holder scheme is then about 50 fs, assuming Gaussian noise distribution.

In Fig. 2(a), two beam FWM data using the two separate lasers are shown, with (solid lines) and without (broken lines) the use of the sample-and-hold switch. The sample was a $GaAs/Al_{0.25}Ga_{0.75}As$ multiple quantum well (30 periods) with well and barrier widths of 100 Å kept at 10 K. The polarization in our experiments was collinear. The sample was chosen so that there is relatively small pulse distortion [11] and the absorption at the continuum is less than 30% of the incident power. Both lasers were tuned such that the HH and the LH exciton energies are within the laser spectra. The spot size at the sample was about 100 $\mu$m, and the densities are approximately $10^{10}$ $cm^{-2}$. Without use of the switch, FWM signal is obviously jitter limited. However, the improved time resolution by use of the switch reveals well defined HH-LH beating. We also note that with mutually perpendicular polarizations, we observe HH-LH beating that is 180° out of phase [12].

Unlike two beams originating from a single laser, the two beams originating separately from our two lasers are mutually incoherent in an average sense; as long as the experiment uses trains of pulses, interference pattern cannot be observed. However, for each pair of pulses from the two lasers, there is basically a fixed phase relation, such that coherent experiments can be performed in the same way as for pulses originating from a single laser. As long as the constant phase shift does not influence the experimental result, such as the HH-LH beating [12], these classes of experiments can be performed with our setup.

In Fig. 2(b), three beam FWM data at 10 K are shown at $\tau_1 = 0$ and varying $\tau_{12}$, with (solid lines) and without (broken lines) the switch. The experimental conditions are roughly the same as in Fig. 2(a). Clear HH-LH beating is observed upon improving the time resolution. In Fig. 3(a), we show the FWM signal as a function $\tau_1$ for a fixed delay of $\tau_{12} = 2$ ps (solid lines), and also as a function $\tau_{12}$ for

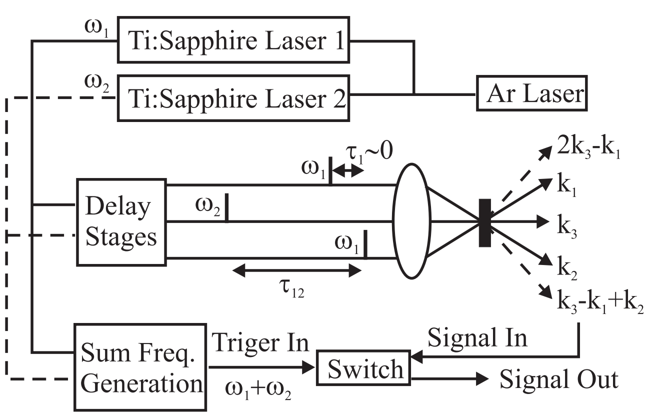


FIG. 1. Schematics of the experimental setup.

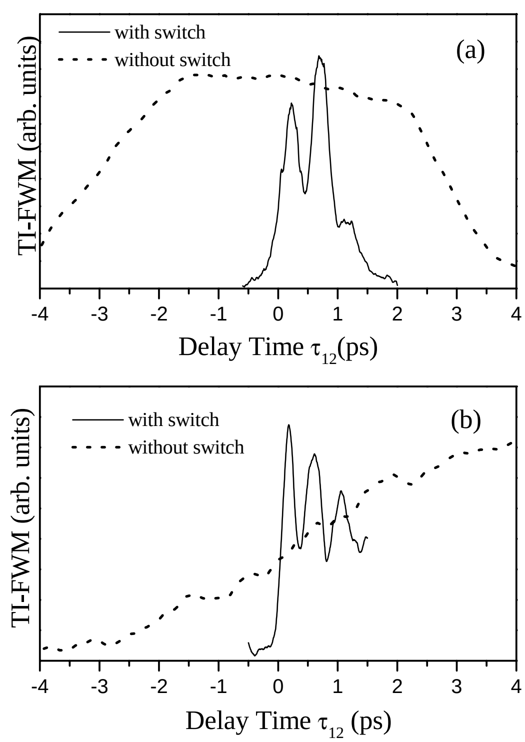


FIG. 2. (a) Two beam FWM signal with (solid lines) and without (broken lines) the switch. Both lasers were tuned near the HH and LH excitons, encompassing both resonances. The sample was a 30 period GaAs/AlGaAs multiple quantum well (100 Å/100 Å $x = 0.25$) kept at 10 K. (b) Three beam, two color FWM signal with (solid lines) and without (broken lines) the switch.

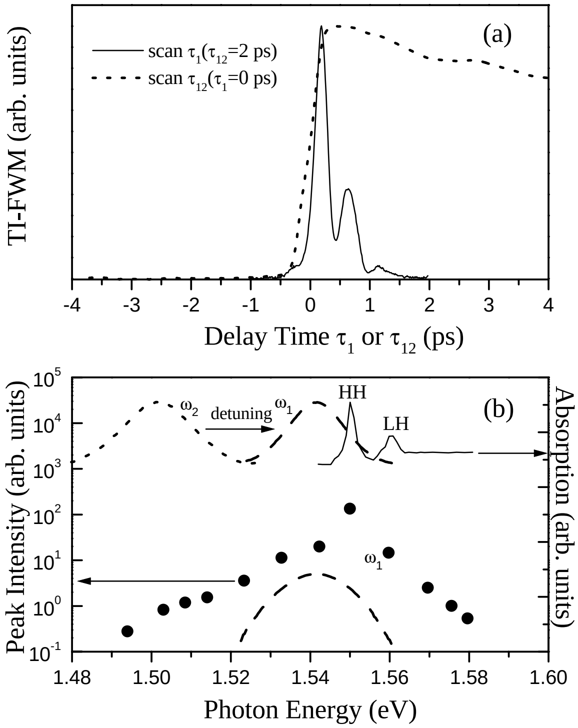


FIG. 3. (a) FWM at 10 K as a function of $\tau_1$ for $\tau_{12} = 10$ ps (solid lines), and $\tau_{12}$ for $\tau_1 = 0$ (broken lines). (b) Peak FWM signal ($\tau_1 \approx 150$ fs) at $\tau_{12} = 10$ ps as a function of the center photon energy of $\omega_2$. $\omega_1$ (broken lines) is fixed slightly below the heavy-hole exciton resonance. The absorption curve is represented by solid lines.

a fixed delay of $\tau_1 = 0$ ps (broken lines). $\omega_1$ is tuned at the HH exciton resonance and $\omega_2$ at completely below the exciton resonances with virtually no absorption (1.52 eV). The HH-LH beating (solid lines) represents the change in the strength of the population grating created by $\omega_1$. Note that there is no HH-LH beating in three beam FWM when we scan $\tau_{12}$, unlike in Fig. 2(b). This is because $\omega_2$ is off resonant.

To investigate this issue further, we have changed $\omega_2$ for a fixed $\omega_1$ [Fig. 3(b)]. $\omega_1$ is tuned slightly below the HH exciton energy, and the peak diffracted signal is plotted as a function of the center frequency of $\omega_2$ at $\omega_{12} = 10$ ps. It is clear that diffraction occurs off the population grating even when there is no absorption in the spectrum of $\omega_2$. On the other hand, the diffraction efficiency is a sensitive function of detuning; there is about a 2 orders of magnitude decrease between the detuning of 10 to 60 meV below the HH exciton. The FWM signal decreases rather quickly as $\omega_2$ is tuned to the continuum. It is surprising that the FWM signal for the continuum is weaker than that for below the band edge for roughly the same detuning. We believe that the destructive interference among the continuum states should be one of the main contributions to this weak signal [13]. In addition, the relatively short dephasing time for the continuum might further decrease the diffraction efficiency [14,15]. A full theory including microscopic dephasing would shed important insight into this interesting phenomenon.

In Fig. 4(a), spectrally resolved (SR) FWM signal at $\tau_1 = 0$ ps and $\tau_{12} = 10$ ps is plotted together with the spectra of the two lasers for various $\omega_2$, for a fixed $\omega_1$ centered approximately at the LH exciton. When $\omega_2$ is at the continuum (top two curves), SR-FWM mostly reproduces the laser spectrum. When $\omega_2$ is completely below the absorption profile (bottom curve), SR-FWM peaks close to the center of $\omega_2$, although its width is somewhat narrower than $\omega_2$. When the laser spectrum is resonant with both the HH and LH excitons (middle curve), collision-broadened HH and LH excitons dominate FWM. In addition, note the existence of separate excitonlike features at the bottom curve. From these results, it is clear that SR-FWM is determined mostly by the laser spectrum of $\omega_2$ off resonance, while the excitonic resonances play an important role when $\omega_2$ is close to them. The possibility of the below band gap, off-resonant SR-FWM signal that does not have much overlap with excitons was first recognized a few years ago by degenerate two beam and three beam FWM experiments [16]. However, the nature of degenerate FWM experiment made it impossible

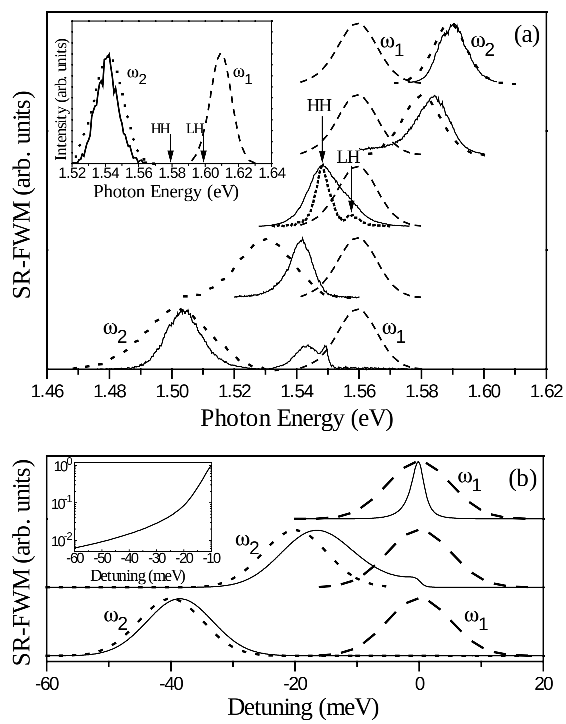


FIG. 4. (a) SR-FWM signal for a fixed $\omega_1$ centered at the LH exciton resonance for various $\omega_2$'s at $\tau_1 = 0$ ps and $\tau_{12} = 10$ ps. In the middle curve, the SR-FWM at lower density is represented by dotted lines, showing clearly defined HH and LH excitons. (Inset) SR-FWM signal for another GaAs/AlGaAs multiple quantum well sample: 30 × (80 Å/80 Å; $x = 0.25$). $\omega_1$ was tuned mostly to the continuum. (b) SR-FWM using a SBE at nearly identical conditions as the experiments. (Inset) Peak FWM signal as a function of detuning below the HH exciton.

to completely separate the off-resonant contribution from its excitonic counterpart. This limitation was a consequence of the necessity for the laser spectrum to have significant overlap with excitonic resonance. Therefore, the maximum detuning was only 8 meV. Our results clearly demonstrate that for $\omega_2$ below the band gap, diffraction of the off-resonant polarization is one of the main contributions to two color three beam FWM. We have verified this in another multiple quantum well sample with a well width of 80 Å as shown in the inset. Here too, the off-resonant SR-FWM is dominated by the laser spectrum.

To analyze our experimental findings we have performed model calculations based on the semiconductor Bloch equations (SBE) [17] projected onto the 1$s$ HH exciton. Dephasing time of 500 fs was introduced phenomenologically. Although it is clear that such a simple model is much too crude to explain all the experimental observations reported here, it can be applied to the off-resonant excitation. The main features of the experiments, especially the presence of strong off-resonant signal and its spectral characteristics, are reproduced well as shown in Fig. 4(b), which used similar conditions as in Fig. 4(a) ($\tau_{12} = 10$ ps, 150 fs pulse width, 15 meV band width). The temporal profile of the off-resonant diffraction follows the laser pulse, as expected from our simple dephasing model. The inset displays the peak FWM signal as a function of detuning, which qualitatively reproduces the experimentally observed decrease of about 2 orders of magnitude between 10 and 60 meV of detuning below the HH exciton. At large detuning, the decrease is Lorentzian-like, as expected from the diffraction of the transient polarization.

In summary, we have performed two beam and three beam two color FWM experiments using two independently tunable femtosecond Ti:Sapphire lasers with 150 fs time resolution, made possible by a simple sample-and-hold switch scheme. HH-LH beating is observed when the jitter is externally compensated. Completely off-resonant FWM signals are observed, and the below the band gap signals are stronger than the signals at the continuum for equivalent detunings. Using the SBE approach, we could reproduce some of the main characteristics of the off-resonant contribution. We believe that our experiments would stimulate further experimental studies, as well as first principle, fully microscopic theoretical investigations.

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