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Mid-Gap Two-Photon Four-Wave Mixing in III-V Semicnductors

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## MIP-GAP TWO-PHOTON FOUR-WAVE MIXING IN III-V SEMICONDUCTORS

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### Abstract

We report the observation of four-wave mixing signals in mid-gap excitation of GaAs and InP, using a femtosecond infrared continuum. We interpret these signals as a two-photon nonlinearity.

## Mid-Gap Two-Photon Four-Wave Mixing in III-V Semiconductors

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Time resolved Four-Wave Mixing (FWM) has been widely implemented in the study of the third order nonlinear optical susceptibility in semiconductors.[1] As a third order process, FWM has been shown to be sensitive to multi-photon excitations, which are difficult to observe with linear optical spectroscopy.[2]. Two-photon (TP) absorption, and more generally multiphoton absorption, have been studied extensively in semiconductors.[3] When a semiconductor is excited below its single-photon bandgap, TP nonlinearities become important. To the best of our knowledge, the contribution of the TP third order susceptibility to FWM signals, which is *not* negligible for below bandgap excitation, has not been previously studied in semiconductors. It has been studied, however, in molecular solutions and polymers.[4]

We report here the observation of FWM signals in GaAs and InP, under mid-gap excitation with femtosecond laser pulses, and demonstrate that the signal is due to TP transitions. We have investigated a 6000 Å GaAs sample, a polished 0.5 mm thick semi-insulating GaAs wafer, and several InGaAs Multiple-Quantum-Well (MQW) samples on InP substrates. The samples were excited by ~150 fsec, "narrow band" ( $\Delta\lambda=30\text{nm}$ ) pulses from a continuum source. The signals were measured in the  $2k_1-k_2$  and  $2k_2-k_1$  configurations, and both time-integrated signals and their power spectra were detected.

Figure 1 shows the time-integrated power spectra, as function of the delay between the exciting pulses, for a 100 Å InGaAs MQW sample on InP substrate, excited just below the one-photon bandgap of the MQW (0.84 eV). Two components can be seen in the signal, which we attribute to excitonic and TP contributions from the MQW and substrate, respectively. We have verified that the TP contribution is always centered near the laser frequency, regardless of it being above or below the one-photon bandgap of the InGaAs

MQW. This component appears even when the MQW is removed from the InP substrate by polishing. On the other hand, only the excitonic contribution remains when the substrate is removed by selective etching. The appearance of both components in Fig. 1 is a verification that both are emitted in the same direction. Similar results were obtained for all samples, both at room temperature and at liquid He temperature.

To prove that this below-gap FWM signal is indeed due to TP processes, we repeated the experiment around the mid-gap energy. Just below the mid-gap of GaAs at liquid He temperature we have measured a sharp decrease of the FWM signal, which cannot be accounted for by the decrease of the continuum intensity in that spectral range.

Within the experimental resolution, the TP signal rises and decays with the autocorrelation time of the pulses. The density dependence of this contribution to the FWM is proportional to  $I^3$  and does not saturate. The dependence on the relative intensities of the exciting pulses, plus a lack of dependence on the crystal orientation, reject alternative explanations which are based on photorefractive and similar effects.

The TP FWM signal we have observed is a demonstration of a strong instantaneous nonlinearity, with no dispersion over a wide frequency range, where linear absorption is negligible.

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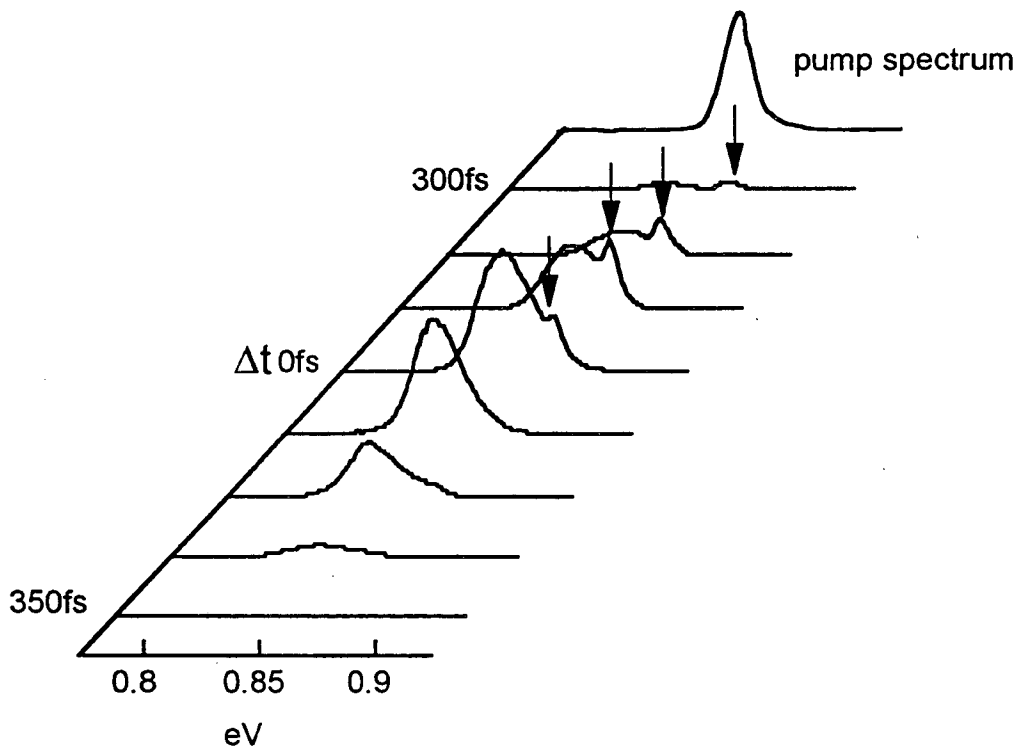
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**Figure Captions**

Figure 1: The time-integrated power spectra, as function of the delay between the exciting pulses, for a 100 Å InGaAs MQW sample on InP substrate, excited just below the one-photon bandgap of the MQW (0.84 eV). The excitonic contribution is on the high energy side (0.87 eV), as shown by arrows.





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