Extreme Nondegenerate Two-Photon Absorption in Semiconductors

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Abstract: Using photon pairs having energy ratios of ~10, gives 2-4 orders of magnitude increases in two-photon absorption in several bulk semiconductors and semiconducting devices (~1 cm/MW in the visible) compared to the frequency degenerate case.

Applications of two-photon absorption (2PA) have been limited, and knowledge of the magnitude of the effect (2PA coefficient $\alpha_2$) has been needed, mainly because it is a limiting factor for applications such as all-optical switching, short wavelength frequency conversion etc. Here we show orders of magnitude increases in 2PA in direct gap semiconductors by using extremely nondegenerate pairs of photons (energy ratios ~ 10/1) that will allow subfemtosecond gated detection, all-optical switching etc. Figure 1 (Left) shows these effects in ZnSe (GaAs, CdTe, ZnO, and ZnS show similar results). We demonstrate gated detection of femtosecond blue light pulses (~390 nm) using femtosecond infrared pulses (~5.6 $\mu$m) in a GaN photodiode in Fig. 1 (right), infrared gated measurements are currently underway and show very promising results. As 2PA is a nearly instantaneous process, this gating is limited only by the gating pulse length (here ~200 fs). A plot of signal vs. time delay between the pulses gives the cross-correlation function while the data for Fig. 1 (Left) are the results of that fitting. The signal here is linear in both the pump energy and 390 nm energy as expected from the 2PA loss given by:

$$\frac{dI_p}{dz} = - (\alpha_1 + 2\alpha_2 I_g) I_p$$

where the $I$'s are irradiances and frequencies of probe (p) and gating (g) pulses respectively, and $\alpha_1$, $\alpha_2 (\omega_p, \omega_g)$ are the one and 2PA coefficients, here indicating that the 2PA is of the probe by the gate. It is also seen in Fig. 1 (Right) that the residual linear absorption of the 390 nm light leads to a signal ~ 3 orders of magnitude smaller than the 2PA signal. In addition, degenerate 2PA of femtosecond, 730 nm pulses of approximately the same pulse energy are nearly 4 orders of magnitude smaller. Note that the 5.6 $\mu$m pump energy is held fixed at 280 nJ while the other energies are varied. This demonstrates the very large 2PA enhancement seen with nondegenerate photons. As an example of how this could be used for all-optical switching, the high Q of an integrated optical ring resonator pumped just below the direct gap could be easily spoiled by a small level of nondegenerate 2PA as in a ZENO effect. Along with the large 2PA is a greatly enhanced nonlinear refraction; however, this falls within the range of large 2PA.

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