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High Intensity Third Harmonic Generation Using a Single Crystal of BBO

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High intensity femtosecond pulses from a CPA laser are used to generate .6 mJ of light at 351 nm (6% efficiency) in a single crystal of BBO. The values of $\chi_{ij}^{(3)}$ are measured for BBO.

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Generation of the third harmonic (THG) in a single process is an idea that has been around since the inception of nonlinear optics.[1–3] However, since the process $\omega + \omega + \omega \rightarrow 3\omega$ is governed by the third order nonlinear electric susceptibility $\chi^{(3)}$, the efficiency of such an interaction has been very low. To this date, the high intensities needed to drive this process has confined most research into higher order harmonic conversion to highly focused beams in gases or liquids. Nonetheless, the increasing number of terawatt-class chirped-pulse amplification (CPA) lasers (and beyond) in recent years has made it possible to achieve very high intensities (> 100 GW/cm^2) in collimated beams without damaging solid material. Using a CPA Ti:sapphire system[7] operating at 1053 nm, we have been able to produce up to .6 mJ of third harmonic light in single crystal of BBO with conversion efficiencies above 6%.

The coupling between third harmonic and fundamental waves is not only due to the third order susceptibility. It has been shown[4–6] that because the fundamental and third harmonic waves are in phase, $\Delta k = 0$, regardless of the intervening processes, it is possible to have significant energy transfer to the third harmonic by two cascaded second order processes. First the second harmonic is generated (SHG) and then is summed with the fundamental to

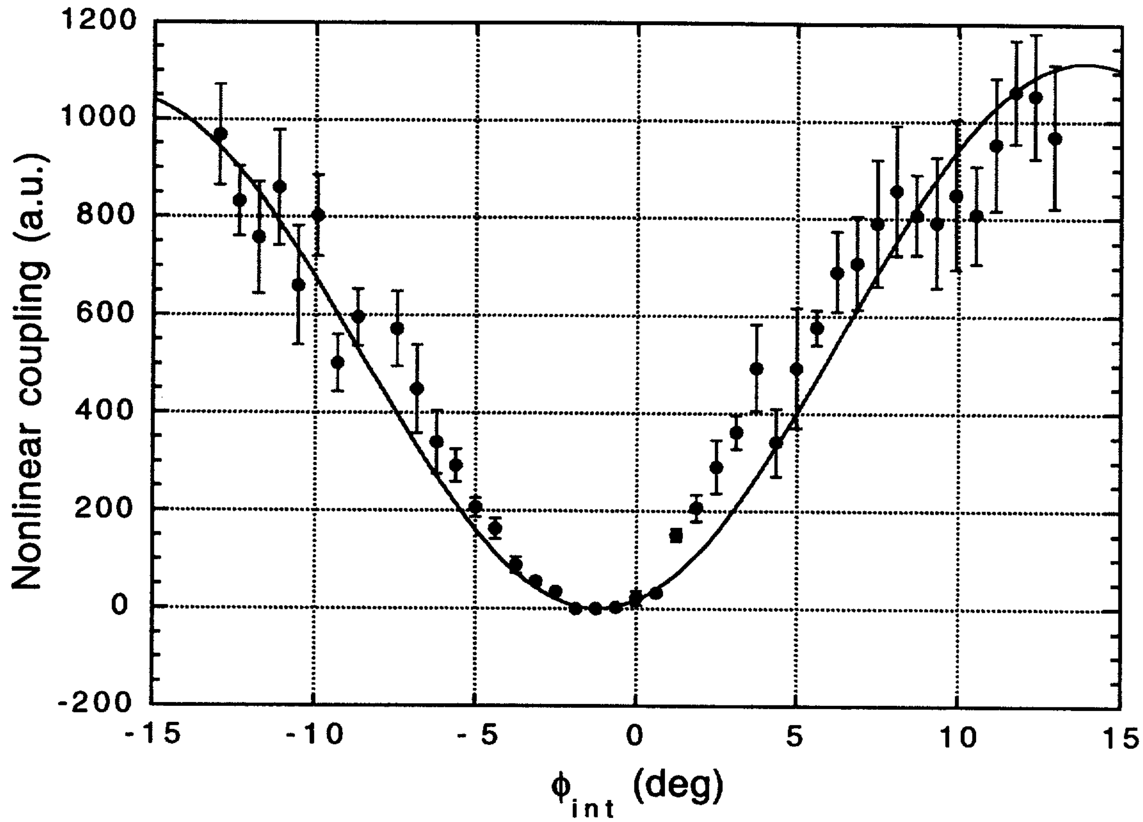

Figure 1: Effective nonlinear coupling for internal azimuthal angle ϕ_{int}.

generate the third harmonic (SFG). This can occur efficiently even though neither the SHG nor the SFG process are phasematched. We were able to measure the THG efficiency for both Type I ($ooo \rightarrow e$, $\theta_m = 37.7°$) and Type II ($ooe \rightarrow e$, $\theta_m = 47.1°$) configurations using the same crystal of BBO (3 mm in length). The effective nonlinear susceptibility χ_{eff} (including both second and third order effects) for Type I phasematching is

$$\chi_{\mathrm{eff}} = A \sin 6\phi + (\sin\theta_m C_{10} - B)\cos 3\phi \tag{1}$$

with

$$A = \frac{2\omega}{c} d_{22}^2 \left[\frac{\cos\theta_m}{n_{2o}\Delta k_{ooe}^{\mathrm{SFG}}} - \frac{\cos^3\theta_m}{n_{2e}\Delta k_{eoe}^{\mathrm{SFG}}} \right] = 77.92\ \mathrm{pm^2/V^2}, \text{ and} \tag{2}$$

$$B = \frac{2\omega}{c} d_{22} d_{15} \left[-\frac{\sin\theta_m}{n_{2o}\Delta k_{ooe}^{\mathrm{SFG}}} - \frac{\cos^2\theta_m \sin\theta_m}{n_{2e}\Delta k_{eoe}^{\mathrm{SFG}}} \right] = -8.76\ \mathrm{pm^2/V^2}. \tag{3}$$

The values used for d_{22} and d_{15} for BBO were 2.2 pm/V and .16 pm/V,[8] respectively. The

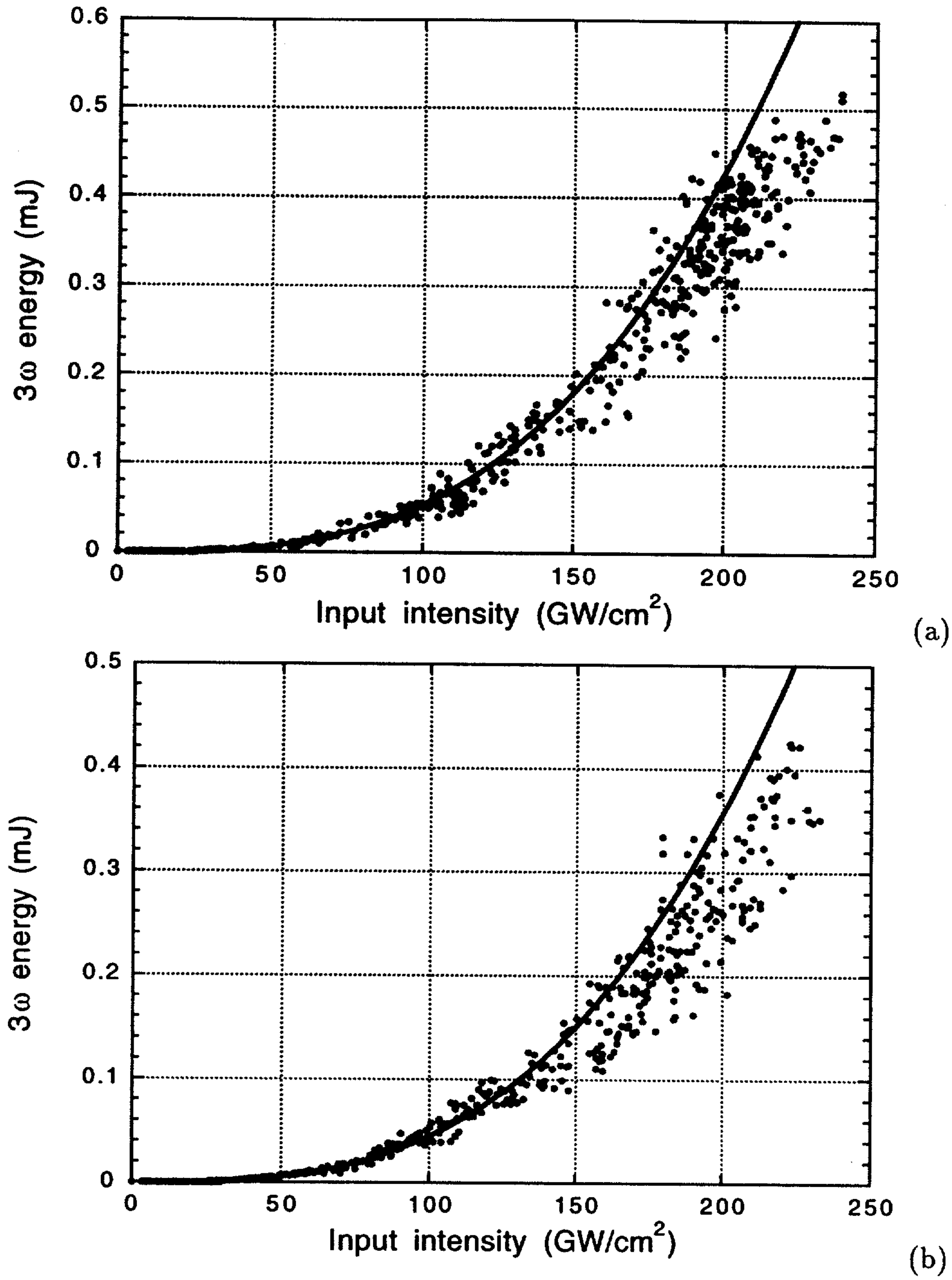

Figure 2: Energy output from single BBO crystal at 351 nm along with cubic fit to low drive points. (a) Type I phasematching with $\phi_{\text{int}} = -15°$. (b) Type II phasematching ($\phi_{\text{int}} = 0$).

nonlinear coupling for Type I THG as a function of ϕ is shown in Fig. 1. Fitting Eq. (1) to the measured data gives a value for $C_{10} = \chi_{10}^{(3)}/4$ of 1.1×10^{-23} m^2/V^2. This compares to an effective value for the cascaded second order coupling of 8×10^{-23} m^2/V^2. A similar calculation and measurement for Type II phasematching yields a value for $C_{11}\cos^2\theta_m + C_{16}\sin^2\theta_m$ of 2×10^{-23} m^2/V^2. This value is only correct to a factor of 2 due to the fact it was not possible to scan ϕ over a large enough range. It is evident that the major contribution to the THG observed is the cascaded process, not the third order process. Finally, the energies at 3ω are shown as a function of input intensities at 1053 nm in Figs. 2(a) (Type I at $\phi = -15°$) and 2(b) (Type 2 at $\phi = 0°$).

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