Quantitative Measurement of the dispersion of third-order nonlinearity in silica in the 1-25 THz range

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We present measurement of the third-order nonlinearity of fused silica responsible for THz-driven second-harmonic generation (TFISH). The measurement is performed by comparing the upconversion signal in traditional Air Biased Coherent Detection (ABCD) with the corresponding detection signal in a solid-state detector based on fused silica (amorphous SiO_2).

ABCD is the conventional method for low-distortion detection of ultrabroadband THz transients [1]. The ABCD method relies on THz-induced second-harmonic upconversion from an intense probe beam, typically at 800 nm wavelength, and thus the absolute value of the third-order nonlinearity of the susceptibility $\chi^{(3)}$ of the gas (typically nitrogen, N_2) determines the absolute intensity of the upconverted SHG signal for detection. Tomasino et al. developed a solid-state equivalent of ABCD (Solid-State Biased Coherent Detection - SSBCD), where the nonlinear medium is a dielectric such as SiO_2 or SiN [2], exploiting the orders-of-magnitude larger nonlinearity of the glass compared to N_2. This results in significantly lower requirements on the intensity of the probe beam and bias voltages used in the heterodyne detection. Here we demonstrate that by careful calibration of the detection conditions (probe intensity, detector bias) it is possible to extract the frequency dependence of $\chi^{(3)}$ across the THz frequency band, here up to 25 THz as shown in Fig. 1.

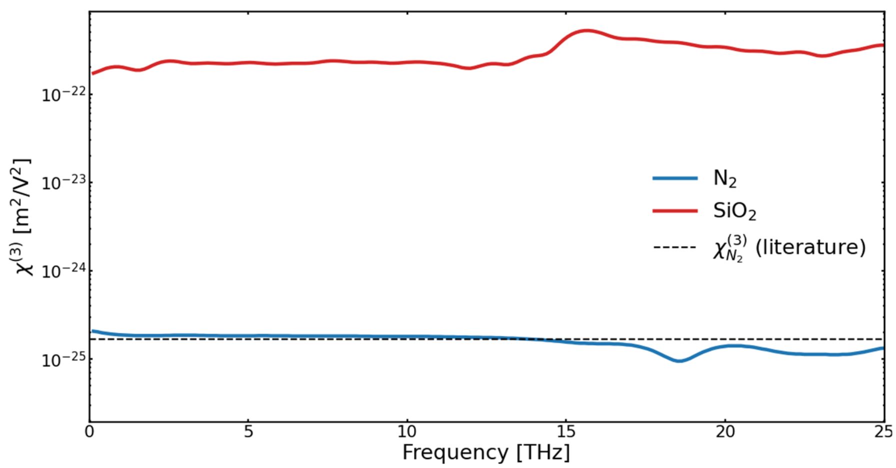

Fig. 1 Extracted $\chi^{(3)}$ for fused silica (red curve) and nitrogen atmosphere (blue curve), compared with the reference value for N_2 (dashed, horizontal line).

For comparison, we also show the apparent $\chi^{(3)}$ of nitrogen gas (ABCD), extracted between two experiments with varying detection conditions. We find a convincing agreement with the literature value for N_2 [3]. Hence, we believe that the reported value for SiO_2 shown in Fig. 1 represents the first measurement of the dispersion of $\chi^{(3)}(\omega)$ across the THz range. We observe the vibrational enhancement of the nonlinearity due to the strong localized phonon mode in SiO2 near 15 THz, confirming the predicted vibrational enhancement of the nonlinearity in SiO_2 [4].

References

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