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*** Withdrawn *** POS-6 Second-harmonic generation in highly dispersive media: Comparison of a new formalism with experimental data

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Second harmonic generation (SHG) is a non-linear optical process where two photons from the pump wave interacting with a non-linear material are combined to generate a new photon, with twice the frequency of the initial ones. The use of organic crystals for SHG is appealing, as the nonlinear coefficients of these crystals are much greater than those of standard inorganic materials, especially when the pump wavelength is close to an optical resonance. However, in such a case, chromatic dispersion must be properly treated.

Indeed, standard formalism for describing the wave equation of SHG in a material has been shown to be flawed in the case of materials with high dispersion. The slowly varying amplitude approximation which is made in the development is not always justified for organic crystals. Solving the equation without this approximation leads to a result with a different amplitude and an additional term, which we refer to as the "zeta term". This term adds an oscillating component in the intensity profile of the second harmonic signal which is not present in standard theory.

Here we experimentally measure the dependence of second harmonic intensity versus pathlength and wavelength in 2-methyl-4-nitroaniline (MNA). Working in photon counting mode, with a motorised translator that allows the sample to be scanned with high reproducibility, we observe what we believe to be a combination of the zeta fringes and an effect caused by multiple reflections, since we are working with thin films. Qualitatively, this agrees with our theoretical predictions and leads us to believe that the standard formalism might lead to significant inaccuracies in the case of highly dispersive materials.

Our results have implications for implementations of non-linear optics in dispersive media, and in the determination of non-linear coefficient with the Maker fringes technique, which relies on the intensity profile of the material.

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