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COMMENT

Comment on 'Optical poling of oligoether acrylate photopolymers doped by stilbene-benzoate derivative chromophores'

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We comment on an earlier paper published in this journal [1]. The authors of that paper report on the all-optical poling of stilbene-benzoate derivative chromophores incorporated into oligoether acrylate photopolymers. The main conclusion that they draw from their study is that, compared with azo-dyes, their materials have better long-lived stability and that the all-optically induced nonlinearity is almost the same as that of the former. They refer, in support of their statement, to a paper (their reference [13]) [2], which was the first one in the literature to report on long-lived all-optical poling in polymers. They ignore later original papers dealing with the same materials and reporting on much larger nonlinearities and long-lived stabilities, as in the earliest reference [2] [3, 4]. It was indeed recognized that the first results in reference [2] were obtained without optimization of the phase [3] in sub-micron thin films and of the intensity ratio between one-and two-photon excitation [4]. More actual results gave a $\chi^{(2)}$ of 150 pm V^{-1} and a dark decay time of 2.5 h for the photo-induced second-harmonic generation (PISHG) signal [4].

But the above is not sufficient to justify a Comment in a scientific journal. The authors describe their results in terms of electric field induced second-harmonic generation (EFISHG): $\chi^{(2)} = \chi^{(3)} E_0$. They estimate a DC field value E_0 in the range 10^2–10^4 V cm^{-1}, which in order to get a $\chi^{(2)}$ close to 2 pm V^{-1} requires a $\chi^{(3)}$ in the range 2×10^{-18}–2×10^{-16} m^2 V^{-2}. This is an unrealistic third-order susceptibility for a dilute transparent organic material (6.5% concentration in weight), out of resonance with the fundamental and harmonic beams: 2–4 orders of magnitude greater than that of CS_2 [5]. Additionally, such an EFISHG effect should be correlated with the third-order microscopic polarizability of the molecules and not the second-order one [4], as claimed in the abstract. A more realistic interpretation emerges obviously from the description of the irradiated region given by the authors themselves: optically polarized measurements reveal that the *so-called induced electric field gratings* have the *said typical narrow size of the cross section in the plane perpendicular to the propagation of the beam (about* 1.2 μm*)*. And they add: *it is somewhat less than the diameter of the incident beam of doubled frequency* .... The cross section of the beams used for PISHG was 60–68 μm at the focal point. Optical physicists would call this phenomenon a 'dark spot' or a 'hole'.

The sample appears burnt by the high fluence used for experiments (around 5.2 GW cm^{-2} fundamental energy). This explains the relative stability of the poling achieved, as well as its relative insensitivity to the intensity used for reading. To finish the Comment, a description of the all optical poling in stilbene-like chromophores was previously delivered by Churikov *et al* [6] (their [4]). Stilbene derivatives usually undergo irreversible *trans–cis* isomerisation upon photo-excitation. The paper commented on only adds confusion to the physics of the process.

I will not discuss any further the surprising aspects of the paper (table 1 for instance), as the main one was discussed above: some of the claimed results are well established, some original. Only the former are exact.

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