Mansoor Sheik-Bahae: contributions to nonlinear optics at CREOL

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ABSTRACT

This is a tribute to Mansoor Shiek-Bahae who passed away July 10, 2023, the first day of the Hawaii Nonlinear Optics conference in Honolulu. See obituaries in Ref. [1]. Mansoor was scheduled to give an invited talk that morning but sent his postdoc Alex Albrecht to give the talk due to his illness.[2] It was a shock for all of us at the conference to have to announce his death there. My presentation at Photonics West will be my third talk about Mansoor's contributions to the field of nonlinear optics. Of course, Mansoor is also well-known for his pioneering contributions to laser cooling which will be outlined by his former student, Denis Seletskiy, in this conference.

Keywords: nonlinear optics, Z-scan, two-photon absorption, nonlinear refraction, second-order cascading

1. INTRODUCTION TO Z-SCAN

Mansoor is perhaps best known for the development of the Z-scan technique for measuring the nonlinear optical properties of materials. As of January, 2024 the first 2 publications on this method have received 2,172 ISI citations (3,170 Google Scholar) [3], and 7,467 ISI citations (10,167 Google Scholar) [4]. The second paper has received the larger number as it gave the mathematical calculations for Gaussian input beams. The work that led up to this involved developing devices to protect sensors including eyes from laser-induced damage. For example, see Fig. 1, where a geometry was used that should be sensitive to both nonlinear absorption and nonlinear refraction and should result in a reduction of the energy and or fluence transmitted by the aperture.[6] We used this geometry as both a nonlinear switch, e.g., when CS_2 was used as the nonlinear medium, but also as a means of screening materials for large nonlinear responses. Increasing nonlinear absorption, NLA, would result in a lower transmittance seen by the detector (D in Fig.1) that has an aperture, A, limiting the area detected. Nonlinear refraction, NLR, would also reduce the transmittance by following a different path to the detector, e.g., dotted line as opposed to the normal linear path, thus reducing the fluence. It was a geometry like this that Mansoor and a student, Ali Said, were using when shining a 10μm TEA pulsed CO_2 laser through a CS_2-filled cuvette that was a few times thicker than the depth of focus. They noticed that the transmittance was very sensitive to the position of the cell in the beam focus. After meeting and discussing, it was realized that this was associated with the NLR. Mansoor proceeded to move the cuvette along the propagation direction and plot the transmittance; thus, the original Z-scan.[3] As it turned out, this was due to the thermal nonlinearity as there was ~0.3 cm^{-1} linear absorption of CS_2 at the CO_2 wavelength. He also altered the geometry to one where the sample (e.g. liquid-filled cuvette) was thin compared to the depth of focus, and then Mansoor could mathematically analyze the results using a Gaussian decomposition method that we had previously used to analyze NLR.[4,5] Mansoor also came up with the name, Z-scan.

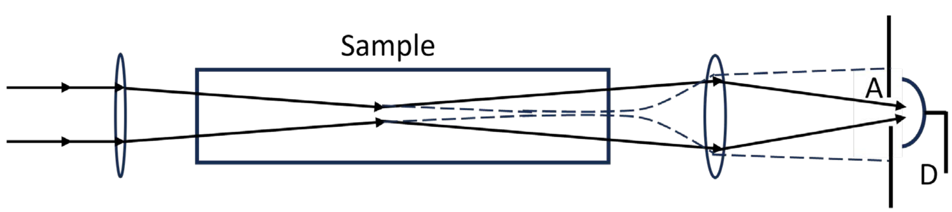

Fig. 1. Geometry for screening materials for large nonlinear responses. Derived from Ref. [6].

2. USES OF Z-SCAN

We immediately used this technique to measure NLR in semiconductors. Specifically, when determining the nonlinear refractive index, n_2, of ZnSe we saw that the bound-electronic n_2 changed from the usual positive value measured at photon energies below the onset of two-photon absorption, 2PA, to negative at photon energies significantly above this energy. This was a clear sign indicating that there should be a similar relation between the real and imaginary parts of the 3rd-order susceptibility, $\chi^{(3)}$, as that between the same properties of $\chi^{(1)}$, i.e., the Kramers-Kronig, KK, relations

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between absorption and index. At this point I discussed this possibility with Mansoor and suggested he try to perform that calculation ignoring all the advice that I had received from multiple physicists that you cannot apply KK to such nonlinear systems. They were correct, as KK relations involve linear dispersion theory. But the way Mansoor got around this was to go to a nondegenerate nonlinearity with two optical beams, excitation and probe, where the equations are linearized. This was a stroke of genius and allowed us to better understand the nonlinear optical properties of multiple materials and classes of materials [7]. One of the ways to argue this point goes as follows. The usual equations for the loss from 2PA (of coefficient α_2) with depth in a material for a single beam is,

$$dI/dz = -\alpha_2(\omega;\omega)I^2, \quad (1)$$

i.e., quadratic in the irradiance, while the corresponding equation for the change in phase of the field is,

$$d\varphi/dz = k_0 n_2(\omega;\omega)I. \quad (2)$$

In the nondegenerate (or 2-beam) case, using an excitation beam (subscript e) and probe beam (subscript p), these equations become:

$$dI_p/dz = -\alpha_2(\omega_p;\omega_e)I_e I_p, \quad (3)$$

which is linear in the probe irradiance, and

$$d\varphi/dz = k_0 n_2(\omega_p;\omega_e)I_e, \quad (4)$$

which is independent of the probe irradiance as are the equations for linear optical interactions. Thus, KK relations can be applied with care. Mansoor also added the nonlinear absorptive processes of Raman and AC-Stark to get the full ultrafast nonlinear refraction [8]. You can think of this as probing a new linear system but changed (dressed) material. The factor of 2 in Eq. 3 simply makes the limit of a single beam, $\omega_p=\omega_e$, correct. There were many publications following this methodology, see, e.g., Ref. [9-11]. Work has continued in this area. For example, his theory showed that using two photons of equal energy gives the minimum 2PA, and if the ratio of photon energies is large, e.g.,10, the 2PA can be enhanced by $\sim 10^3$. This enhancement has been used in an uncooled GaN photodiode for sensitive, gated IR detection.[12] Similar work is continuing elsewhere, see for example Ref. [13].

I will just relate one more of Mansoor's contributions during his 7+ years he spent at CREOL and that involves his contributions to creating the field of cascaded second-order nonlinear optics. Mansoor was working with our student Richard Desalvo on measuring the n_2 of the second-harmonic generating, SHG, crystal, KTP. Richard measured a different value of n_2 nearly every day. Eventually after completely rebuilding his experiment, he measured a different sign. After a group meeting that included all the authors on the publication, Ref. [14] we considered microscopic cascading applied to SHG [15]. Mansoor was the first to show me the mathematical solution for how a frequency-doubling crystal leads to both loss of the fundamental, mimicking 2PA, but also how this leads to NLR off phase match. This was the start of considerable research into macroscopic cascading of second-order nonlinearities. Fig. 2 shows the results of a Web of Science search for this topic. Note that this history begins in 1992 coinciding with our publication, Ref. [14]. As the figure shows, this field is still bearing fruit.

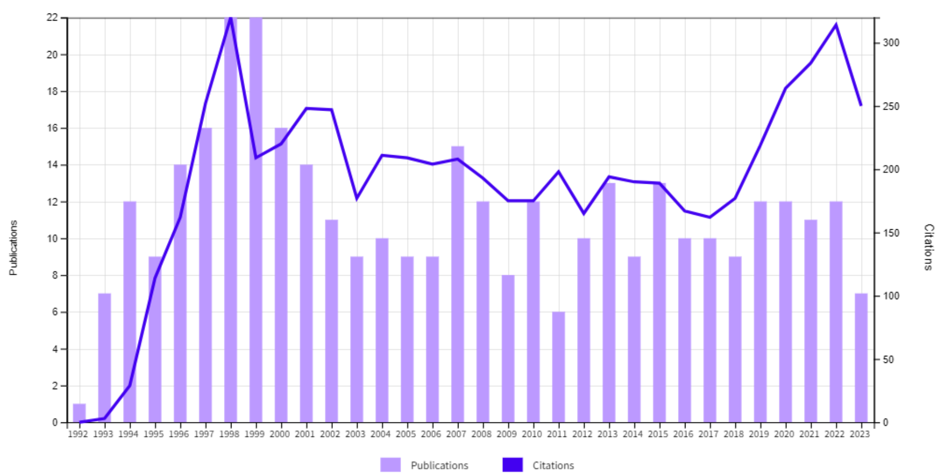

Fig. 2. Results for the number of citations vs. year for the topic of cascaded second-order nonlinearities from the Web of Science.

Finally, on a personal note, I was greatly honored to share the 2012 R. W. Wood Prize of Optica (formerly OSA) with Mansoor for the invention and application of his Z-scan technique.

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