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# Saturation of the all-optical Kerr effect in solids


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**Abstract**

We discuss the influence of the higher-order Kerr effect (HOKE) in wide band gap solids at extreme intensities below the onset of optically induced damage. Using different theoretical models, we employ multiphoton absorption rates to compute the nonlinear refractive index by a Kramers-Kronig transform. Within this theoretical framework we provide an estimate for the appearance of significant deviations from the standard optical Kerr effect predicting a linear index change with intensity. We discuss the role of the observed saturation behavior in practically relevant situations, including Kerr lens mode-locking and supercontinuum generation in photonic crystal fibers. Furthermore we present experimental data from a multi-wave mixing experiment in $BaF_2$ which can be explained by the appearance of the HOKE.

Nonlinear optics frequently uses a perturbative expansion of the dependence of the polarization $P$ on the external electric field $E$, which, for centrosymmetric materials, can be written as $P = \epsilon_0(\chi^{(1)}E + \chi^{(3)}E^3 + \dots)$. This expansion is typically carried out to third order, with orders five and higher being neglected. This simple perturbative picture breaks down when intensities approach the tunnel regime, and a multitude of harmonics may appear in the process of high-harmonic generation (HHG [1]). While this process is well understood and forms the foundation of attosecond physics [2], much less is known about the self-refraction counterpart of HHG, i.e., the higher-order Kerr effect (HOKE). Recently, experimental results [3] on the appearance of the HOKE in argon and air constituents were presented. This report on Kerr saturation initiated a controversial debate on possible consequences for our understanding of filamentation [4]. In a previous publication [5] we introduced a formalism for predicting the onset of significant HOKE contributions from the theoretical side. Here we extend this formalism to solid dielectric materials.

So far, only isolated reports on high-order nonlinearities in solid materials exist, see, e.g. [6]. High harmonics and multiphoton absorption effects are typically only discussed in the context of optical damage induced by femtosecond pulses [7]. A significant self-refraction counterpart of these effects could impact our understanding of femtosecond pulse generation methods like Kerr lens mode-locking (KLM). In fact, it appears puzzling that KLM is essentially limited to the generation of pulses with less than 1 MW peak power. Another possible candidate is spectral broadening in photonic crystal fibers, which tends to saturate while increasing the input power beyond a certain level. Additionally, we routinely observe indications for a possible role of higher-order effects in multi-wave mixing experiments with amplified pulses, see the example using $BaF_2$ as nonlinear medium in Fig.1 which are indicative of higher-order processes, corresponding to nonlinear susceptibilities up to $\chi^{(13)}$ if caused by the HOKE.

To shed some light on the possible role of the HOKE in dielectric solids, we applied several proven models for multiphoton absorption, calculating the nonlinear refractive index via Kramers-Kronig transform as well as the plasma contribution predicted by Drude theory. Among these models is the original Keldysh theory [8] which is based on first order perturbation theory with specially dressed wave functions including the effect of the external electric field. Moreover, we use a similar model of Brandi and de Araújo [9] based on an $S$-matrix approach and, finally, the second-order perturbation theories of Wherrett [10] and Sheik-Bahae *et al.* [11]. As a first test, a comparison of the resulting two-photon absorption (TPA) coefficients in $SiO_2$ is

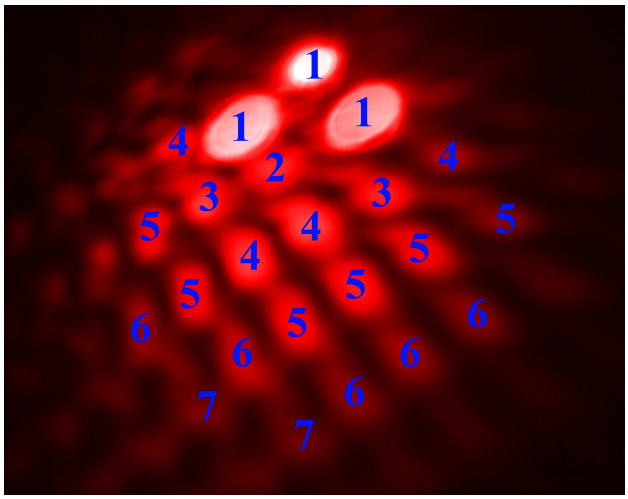


Figure 1: (color online) Beam pattern observed in the far field of a multi-wave mixing experiment in a 500 $\mu$m thick $BaF_2$ crystal at elevated intensities, i.e., ≈10 TW/cm$^2$ in a single input beam with 800 nm center wavelength, 10 fs pulse duration and peak power of 40 MW at 3 kHz repetition rate. The experiment is carried out in a boxcar geometry with a focal length of 20 cm and a crossing angle of ≈2°. Three primary input waves have been tagged by '1', secondary waves with numerals according to order.

shown in Fig. 2(a). With the exception of the original Keldysh theory, all these theories agree within some $10\%$. From this data, we compute Kramers-Kronig transforms of the degenerate TPA coefficients $\beta_2$ as proposed in [11]

$$n_2(\omega) = \frac{c}{\pi}\int_0^\infty \frac{\beta_2(\frac{1}{2}(\omega+\Omega))}{\Omega^2-\omega^2}d\Omega, \tag{1}$$

where $c$ is the speed of light and $\omega$ the laser frequency. This formalism yields the dispersion curves of the nonlinear refractive index $n_2$ as shown in Fig. 2(b). Again, it is striking that the models of Brandi, Wherett, and Sheik-Bahae all yield excellent agreement with measured data [12], while direct application of the Keldysh theory appears to underestimate nonlinear refraction by about a factor 2. Despite quantitative differences, all curves share the same prototypical behavior of positive nonlinear refraction up to a peak at about half the band gap energy with subsequent sign reversal of the effect, consistent with experimental data. Increasing the order $k$ of the multiphoton absorption $\beta_k$, this sign reversal appears increasingly shifted toward the infrared until the corresponding nonlinear index $n_{2(k-1)}$ eventually becomes negative at the laser wavelength. The same observation was made in [5] for noble gases, where it was found that for sufficiently high intensities the total nonlinear refractive index starts to saturate and finally becomes negative. In the following, we chose the Brandi theory [9] for computation of the HOKE. The materials under investigation are $SiO_2$ and $Al_2O_3$, i.e., two technically highly relevant materials, and the alkali halides $BaF_2$ and LiF showing an even higher band gap.

Bearing the above considerations in mind, it is not in question whether Kerr saturation is observed for any of these materials, but whether the saturation is observed before or after index changes due to plasma formation become significant. To address this question, we applied the identical MPA model by Brandi to calculate the formation of plasma according to the rate equa-

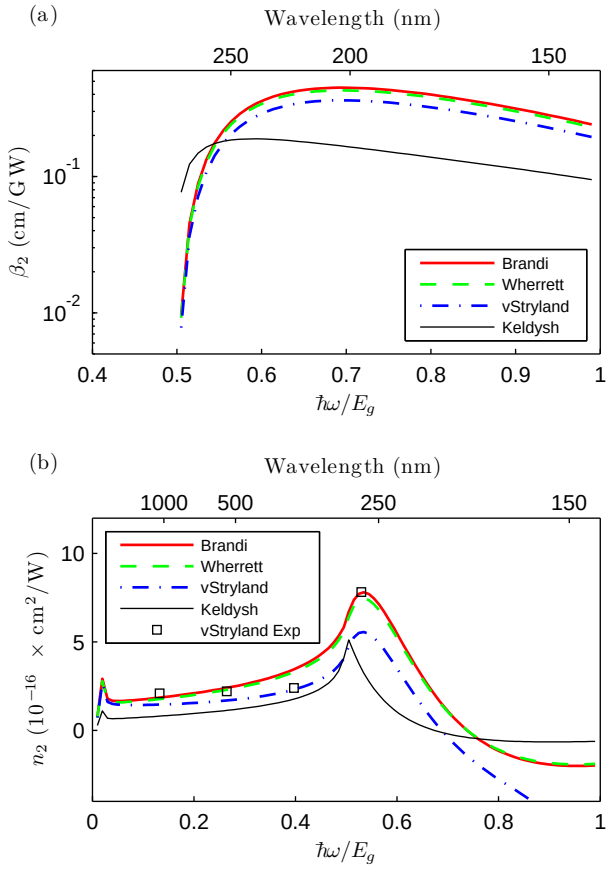


Figure 2: (color online) (a) Two-photon absorption coefficients for $SiO_2$. (b) Theoretically calculated [8, 9, 10, 11] and experimentally measured dispersion [12] of the nonlinear refractive index $n_2$ for $SiO_2$.

tion from [7]

$$\frac{d\rho(t)}{dt} = \sum_k \beta_k I(t)^k + \alpha\rho(t)I(t) - \frac{\rho(t)}{T_{rec}}, \tag{2}$$

where the first dominant term sums over all possible multiphoton processes, the second term describes avalanche ionization, and the last and rather negligible term accounts for recombination. The time-dependent plasma density is computed by integrating (2), assuming a 10 fs Gaussian pulse. The parameters $\alpha$ and $T_{rec}$ are extracted from [7], and multiphoton absorption up to $k = 15$ is considered. The change of the index of refraction due to plasma is evaluated according to Drude theory

$$\Delta n = -n_0 + \sqrt{n_0^2 - \rho/\rho_c}, \tag{3}$$

where $n_0$ is the linear refractive index and $\rho_c = \omega^2 m_e \epsilon_0 / q_e^2$. The critical plasma density for material damage is on the order of $10^{21}$ cm$^{-3}$.

Figure 3 shows a direct comparison between the resulting Kerr effect and the plasma contribution to the refractive index. Both effects are evaluated at the peak of the 10 fs Gaussian pulse. For all four cases investigated, one observes a remarkably similar behavior, with Kerr saturation kicking in at an index change of about $2 - 2.5 \times 10^{-3}$ and plasma clamping appearing at

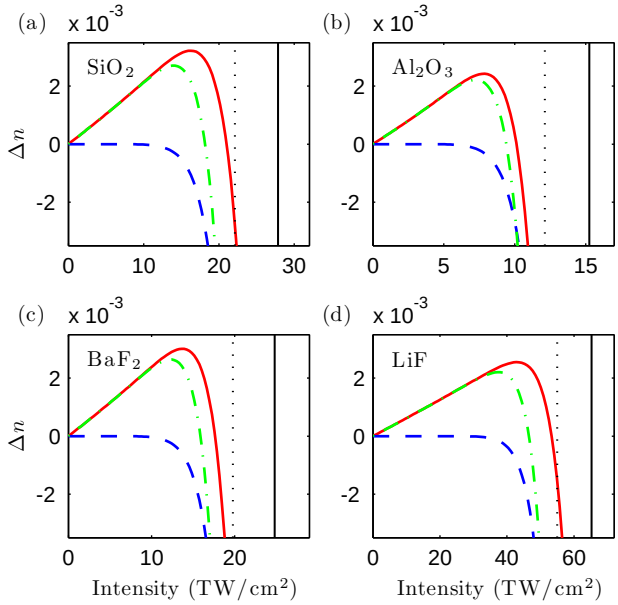


Figure 3: (color online) Contributions to the refractive index from the Kerr-effect (solid red line), plasma formation (dashed blue line) and their sum (dash-dotted green line) at the center of a 10 fs Gaussian pulse as a function of peak intensity for (a) $SiO_2$ (b) $Al_2O_3$ (c) $BaF_2$ and (d) LiF. The dotted and solid vertical lines indicate a plasma density of $10^{20}$ cm$^{-3}$ and $10^{21}$ cm$^{-3}$, respectively, marking the range in which damage is expected.

intensities slightly below Kerr saturation. Note that the maximum index change for noble gases reported in [5] is several orders of magnitude smaller and is achieved at even higher intensities, i.e., $\Delta n = 5 \times 10^{-7}$ at $112$ TW/cm$^{-2}$ for He and $\Delta n = 2 \times 10^{-5}$ at $30$ TW/cm$^{-2}$ for Xe. However, the process in solids is dominated by plasma clamping, yet with Kerr saturation enhancing the negative index contribution by up to a factor two, as in the case of $Al_2O_3$. It is important to understand that both these effects do appear well below the damage threshold for 10 fs pulses. A resulting plasma density of $10^{21}$ cm$^{-3}$ is normally considered a criterion for inevitable optical damage [7], as indicated by a vertical solid black line in Fig. 3. To exclude even occasional optical damage, we included a second dotted black line in the figures indicating a plasma density of $10^{20}$ cm$^{-3}$. Quite clearly, plasma and Kerr saturation appear concomitantly below this reduced damage threshold, indicating inextricably combined action of both effects for filament formation in solids.

While it is now clear that Kerr saturation has an amplifying effect in the formation of filaments in solids, there are other interesting situations, e.g., KLM in Ti:sapphire lasers. Here, one typically assumes maximum nonlinear phase shifts of about $\pi$ per passage through the crystal. Assuming $n_2 = 3.5 \times 10^{-16}$ cm$^2$/W and a crystal length of $2.5$ mm, this translates into an intracavity intensity of about 0.5 TW/cm$^2$, which is clearly insufficient for expecting Kerr saturation in this scenario. At the input facet of a photonic crystal fiber, however, such effects become non-negligible in the extreme case of launching a 7 fs pulse with 5 nJ energy into a fiber with 2 $\mu$m core diameter. These parameters are readily obtainable with commercially available technology, yielding an intensity of 30 TW/cm$^2$ that may play a decisive role in the first few millimeters of propagation inside the photonic crystal fiber [13]. Finally, moving to $BaF_2$, we compute a saturation intensity of 13 TW/cm$^2$. According to our simulations, therefore, we believe that the findings

in Fig. 1 are due to the combined action of a plasma grating [14] and Kerr saturation, with significant plasma densities augmented by a reduction of the refractive contribution from bound electrons. The HOKE therefore already appears to play a non-negligible role in this scenario.

In conclusion, we believe that both, our experimental findings as well as our theoretical analysis shed new light on higher-order nonlinearities in optics. While their role in harmonic generation is certainly undisputed, the self-refraction counterpart of these $\chi^{(n)}$ $(n > 3)$ effects is much more difficult to separate from low-order nonlinearities, as all these effects appear at the same wavelength. However, the role of HOKE contributions in solids at 800 nm appears significantly less pronounced than reported for the noble gases. In the near-infrared, therefore, the appearance of the HOKE in solids goes hand in hand with plasma formation. However, our simulations also indicate a lesser role of plasma effects if a significantly larger number of photons is required to reach the bandgap. In the mid-infrared in crystalline solids, filamentation may appear without dissipative multiphoton absorption effects [15]. Nevertheless, the HOKE appears to have a subtle and often underestimated role in nonlinear optics.

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