SAND2012-1574C

Measurement of nonlinear refractive index and MPI coefficients in gases using a wavefront sensor

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HIGH INTENSITY LASERS AND HIGH FIELD PHENOMENA (HILAS)

Berlin, Germany March 19 – March 21

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

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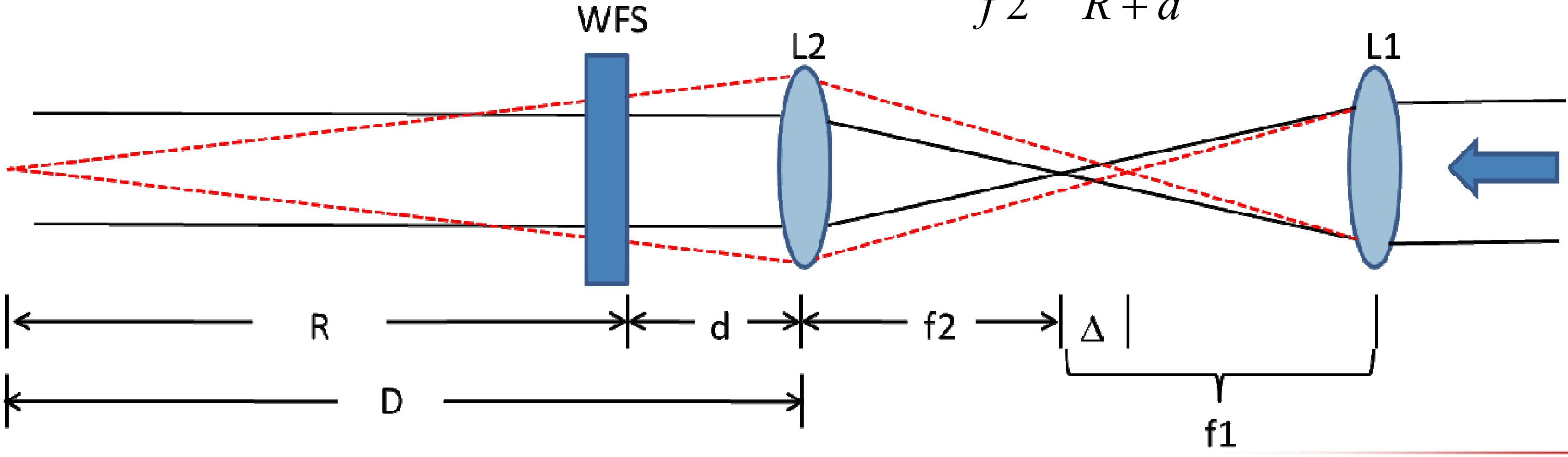
Motivation

- **Many nonlinear optics phenomena are due to the Kerr effect and its associated nonlinear refractive index n_2.**
- **A good measurement of n_2 is therefore vital for understanding these phenomena, such as: self-focusing, self-phase modulation, spectral broadening, self-compression, and filamentation.**
- **At our wavelength and pulsewidth of interest (1054 nm, 540 fs) no such measurements for n_2 exist.**
- **Using a wavefront sensor we propose to directly measure the self-focusing contribution in a focused beam geometry setup.**

Conceptual Experimental Setup

- Initially, a collimated low energy beam is focused (L1=f1) into a gas cell and re-collimated with L2=f2.
- As the laser beam power increases, the focus will move a distance Δ toward L1. This new focus is then re-imaged by L2 at a distance R+d, where R is the radius of curvature measured at the wavefront sensor.
- Δ can then be calculated from the thin lens equation:

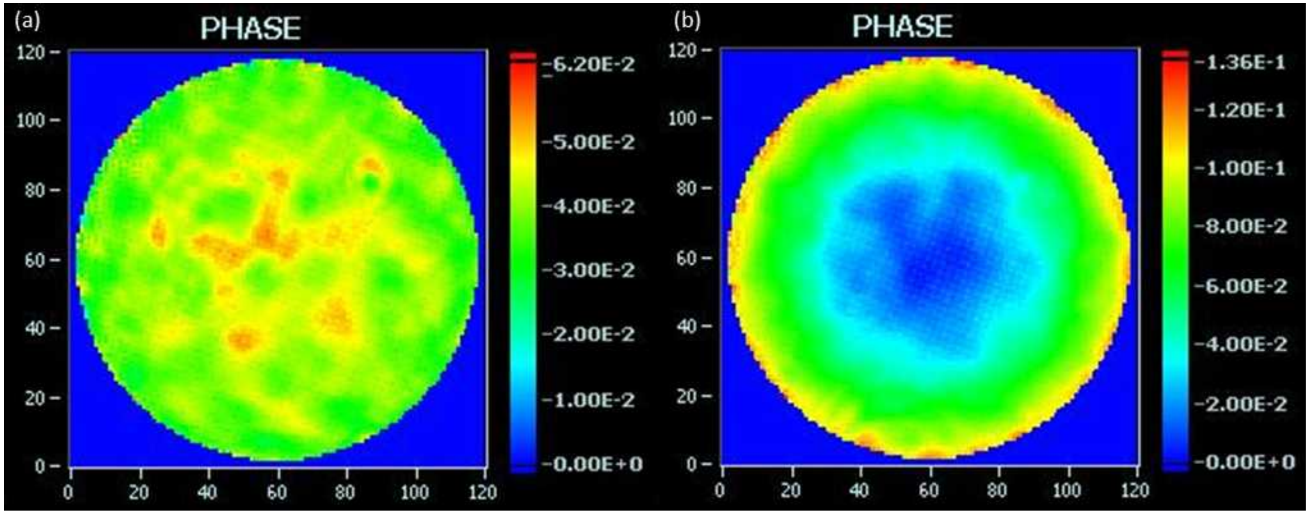
$$\Delta = \frac{1}{\frac{1}{f2} - \frac{1}{R+d}} - f2$$



Wavefront Sensor Data

- Typical measurement from our Phasics SID-4 wavefront sensor:
  - (a) At 0.3 mJ beam energy, a flat wavefront with a PV "defocus" term of -0.005 waves is recorded.
  - (b) At 5.4 mJ of energy a "defocus" term of 0.05 waves is detected.
- The radius of curvature R can be calculated from the Zernike "defocus" term Z^0_2. For a WFS pupil of radius a:

$$R = \frac{a^2}{4 Z_2^0 \lambda}$$



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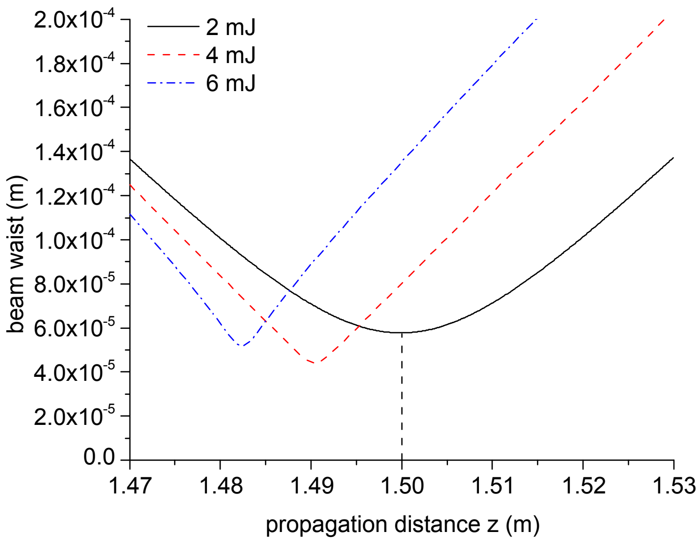
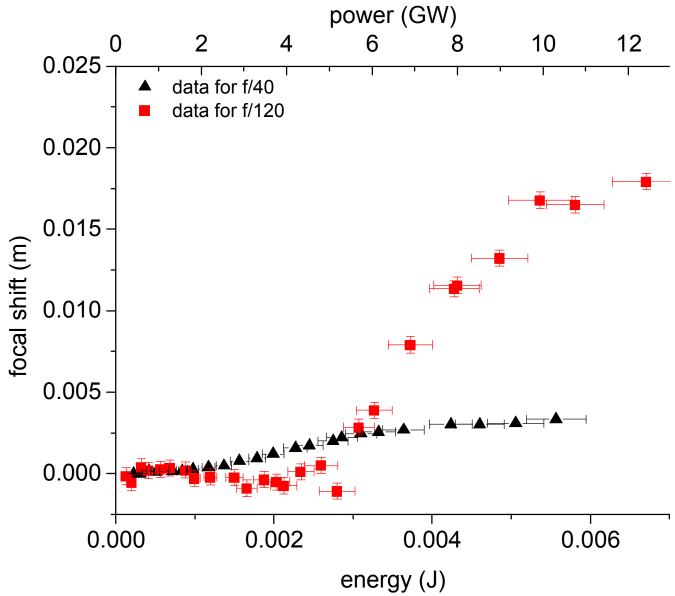
Nonlinear Focal Shift

- **In a focusing beam geometry, to first order, nonlinear focal shift is due to a combination of:**
  - **Kerr effect: As the beam power increases, the Kerr effect leads to self-focusing through the intensity dependent index change $\Delta n = n_2 I$.**
  - **Ionization: As the beam intensity increases near focus, the laser generated electron density will de-focus the beam before it reaches the geometrical focus. This also leads to a focal shift toward the input optic L1.**



Laser beam waist versus propagation distance. Note how the waist shifts toward the input optic as the beam power increases.



Measurement showing focal shift versus laser energy/power. Note the distinctly different behaviors for the two different focusing geometries.

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Theoretical Modeling

- The spatial evolution of the beam waist *w(z,t)* along the propagation axis *z* is governed by the Kerr effect (and its associated n_2) as well as ionization (and its corresponding ionization coefficient $\sigma^{(K)}$).

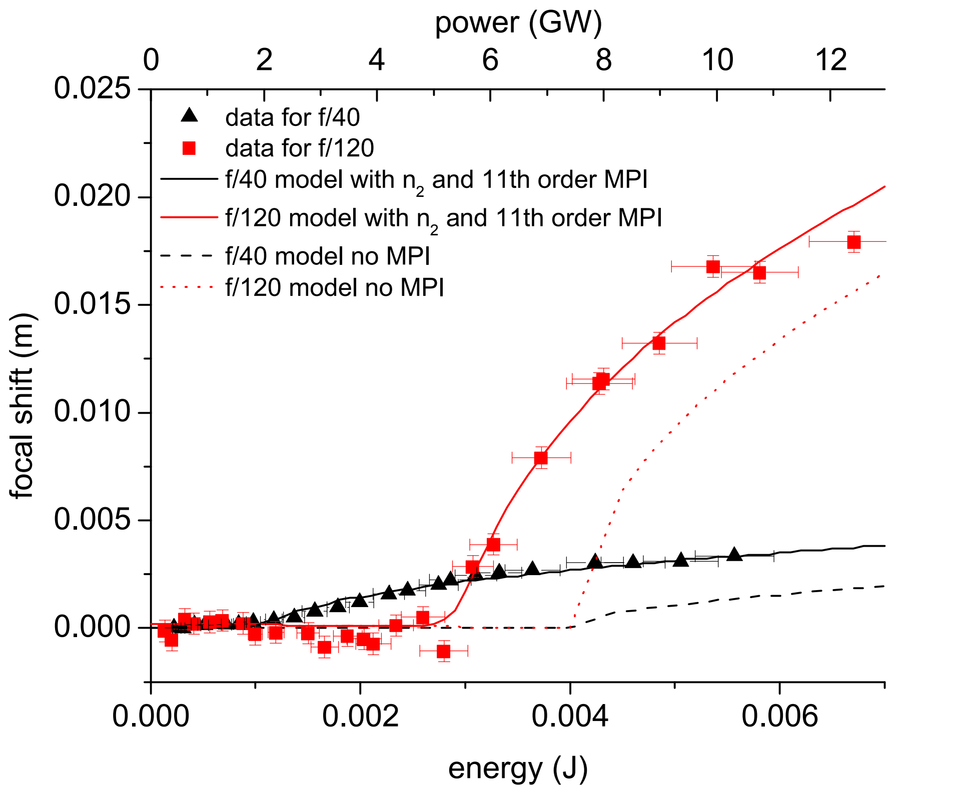
$$\frac{\partial^2 w(z,t)}{\partial z^2} = \frac{4}{k^2 w(z,t)^3}\left(1 - \frac{P(t)}{P_{crit}}\right) + \frac{2K}{(K+1)^2 n_0 N_{crit} w(z,t)} \times N_e(z,t)$$

- The first two terms describe diffraction and self-focusing and the last term represents a Kth order multi-photon ionization (MPI) process.
- For a square temporal pulse profile one can obtain a simple equation that is solely dependent on propagation distance z.

$$\frac{\partial^2 w(z)}{\partial z^2} = \frac{4}{k^2 w(z)^3}\left(1 - \frac{P_0}{P_{crit}}\right) + \frac{2K N_{O_2}\left(1 - e^{-\sigma^{(K)} I_0^K \left(\frac{w_i}{w(z)}\right)^{2K} \tau}\right)}{(K+1)^2 n_0 N_{crit} w(z)}$$

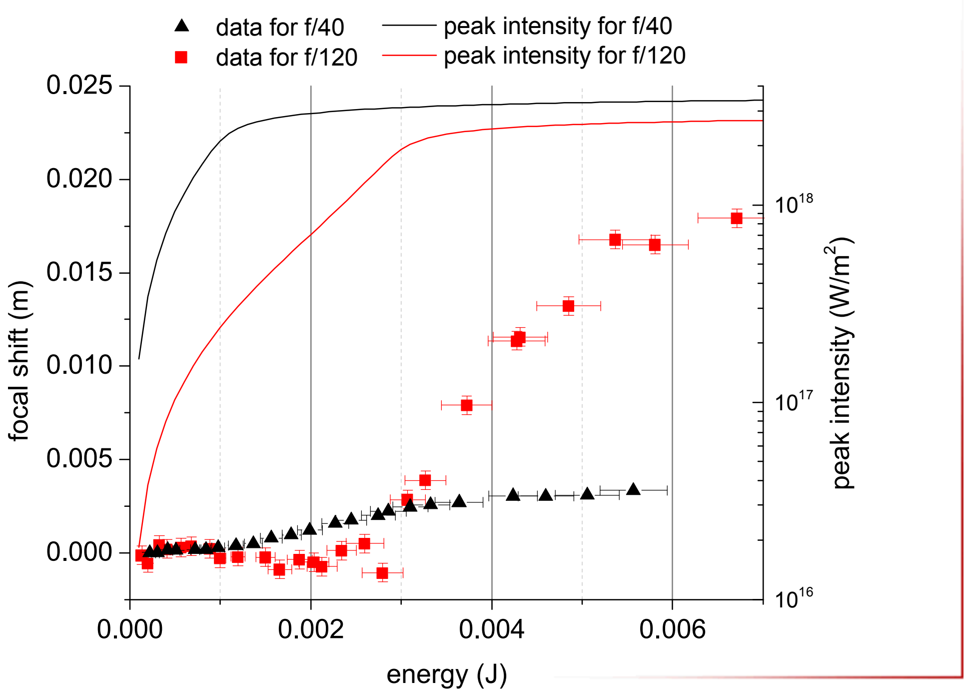
Experimental Results

- The above equation can be numerically integrated using initial conditions: $w_i = w(z=0)$ and $dw(0)/dz = -w_i/f1$.
- Experimental data can then be fitted by varying n_2 and $\sigma^{(K)}$ (K=11 for air/oxygen) until both focusing geometries can be reproduced satisfactory.
- Since there exist two data sets for two unknowns, one can extract a unique solution.
- For our data set:
  - $n_2=(2.6\pm0.2)\times10^{-23}$ m^2/W
  - $\sigma^{(11)}=(3\pm1.5)\times10^{-191}$ m^{22}W^{-11}s^{-1}



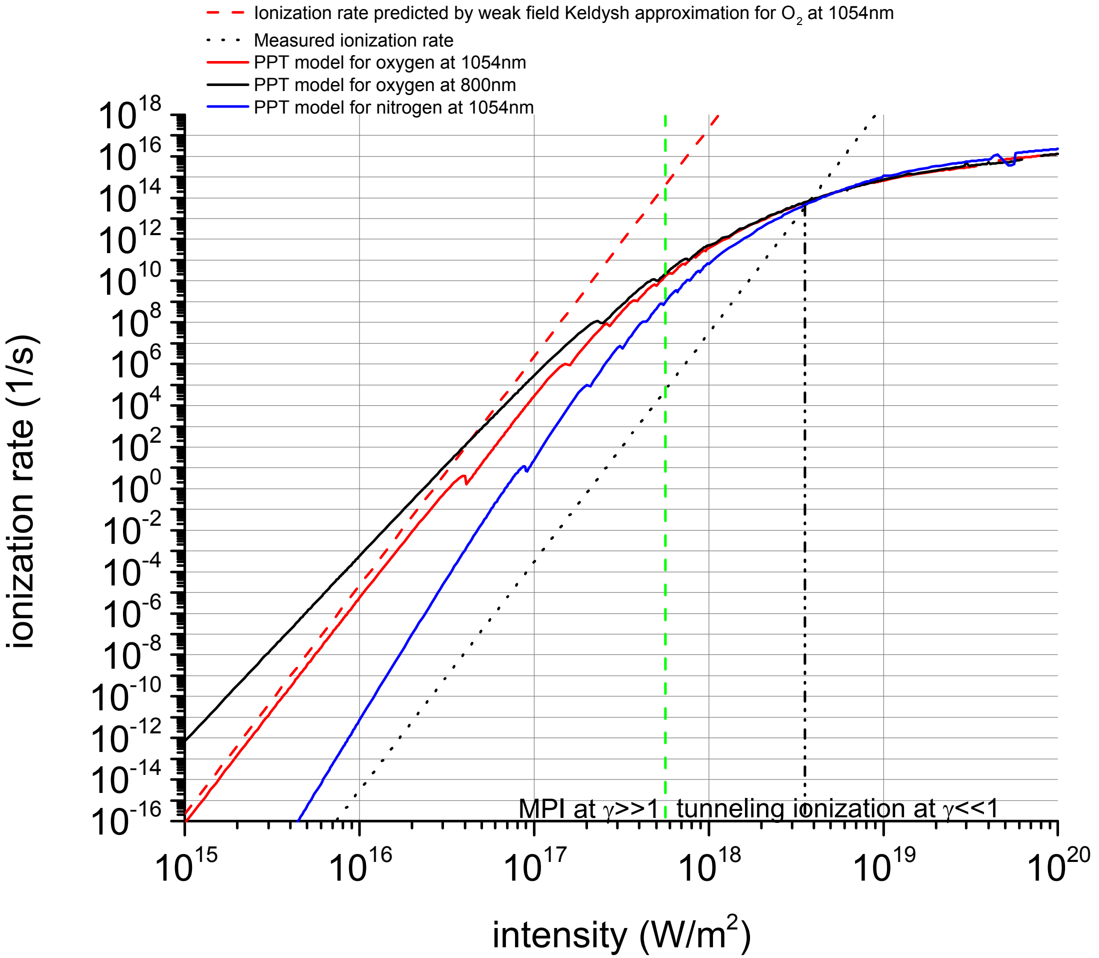
Data Interpretation

- The measured n_2 value compares well with published data at wavelengths close to 1054 nm.
- However, $\sigma^{(11)}$ is about 10 orders of magnitude lower than would be expected from the weak field Keldysh approximation. The reason for that is the fact that the experiment operates in the intensity clamped regime of 3.5×10^{18} W/m^2 which is tunneling ionization dominated.
- To get a better understanding of the ionization dynamics, we have simulated ionization rates for different gases using the PPT model.



Data Interpretation

- **PPT model of gas ionization rates versus laser intensity.**



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Conclusion

- We have demonstrated a novel method for measuring n_2 and the associated ionization rate under atmospheric conditions.
- It has been shown that ionization plays a large role in the self-focusing behavior of ultra-intense laser beams.
- A semi-analytical model has been employed to account for Kerr self-focusing and plasma de-focusing. This model reproduces the data well and allows for extraction of n_2 measurements and ionization rates.
- The measured ionization rates are consistent with predictions from the PPT theory.
- In principle, the above technique is applicable to a broader range of wavelengths and pressures. However, one should make some initial estimates of the expected focal shift in order to verify that the wavefront sensor is sensitive enough to register the expected wavefront deformations.