# Bound Electron Nonlinearity Beyond the Ionization Threshold

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**Abstract:** We measure the residual bound electron nonlinear polarizability well above the ionization threshold in a range of gases, and find that it scales approximately linearly with laser intensity.

**OCIS codes:** (020.4180) Multiphoton processes; (260.3230) Ionization; (190.3270) Kerr effect

Quantitative understanding and accurate simulations of femtosecond filamentation require a well understood optical nonlinearity, including the optical Kerr effect and the ionization induced plasma response. In the noble gases and air, the Kerr effect, which includes bound electronic, rotational, and vibrational contributions [1,2], has been carefully measured. In contrast, knowledge of the ionization-induced nonlinearity relies on relatively indirect measurements, performed in very low pressure gas samples [3,4], which have no time resolution. In addition, there remain persistent questions about the nature of the nonlinearity at ionizing intensities: is there a significant higher-order Kerr effect [5]? Here, we present all-optical absolute measurements of the optical nonlinearity in Ar, Kr, Xe, $N_2$, and $O_2$ at intensities ranging from well below to well above the ionization threshold.

Our measurement technique, which uses chirped supercontinuum pulses to probe the nonlinear phase shift induced by a pump pulse as both propagate through a thin gas target [6], has recently been updated to yield 2D+1 nonlinear phase shift information (2D transverse space plus time). Typical results are shown in Fig. 1. We observe a positive phase shift during the early portion of the pump pulse due to the Kerr effect and an accumulating negative response from ionization.

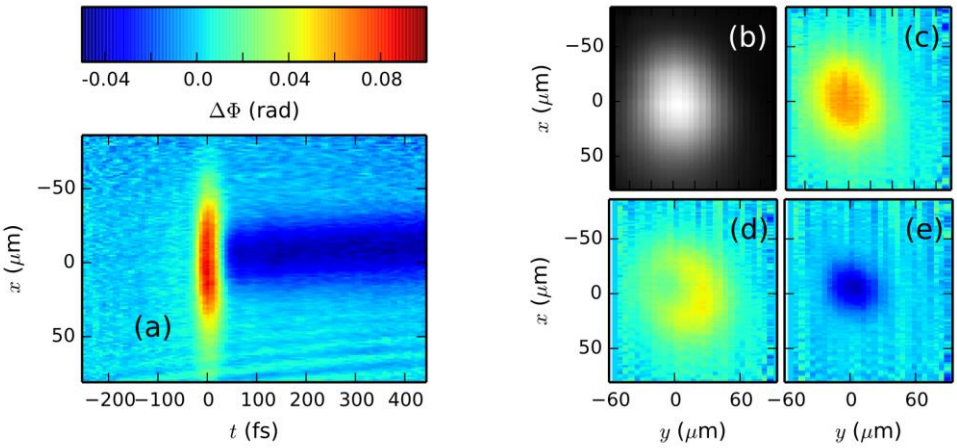


Fig. 1. Experimental results in Ar for peak pump intensity of 95 TW/cm$^2$ and pulsewidth 42 fs. (a) Measured spatiotemporal phase shift $\Delta\Phi(x,y_0,t)$. (b) An image of the pump spot at the gas target. (c) The phase shift $\Delta\Phi(x,y,t = -14$ fs), showing mostly the Kerr response. (d) Phase shift $\Delta\Phi(x,y,t = +25$ fs), showing the Kerr response on the wings and the growing plasma contribution in the center of the beam. (e) Phase shift $\Delta\Phi(x,y,t = +100$ fs), showing the dominant plasma contribution after the pump pulse. The peak of the pump pulse defines zero for the $x$, $y$, and $t$ coordinates.

Our results are qualitatively similar to what we have reported previously at high intensities [2]. However, those measurements showed effects from pump and probe refraction, and degraded time resolution [6]. Here, we use a larger (x2.5) pump spot, we have optimized the time resolution by reducing the probe chirp and improving the spectral resolution [6], and we measure the full 2D+1 phase shift by measuring the phase shift across the entire probe spot. The latter allows us to Fresnel propagate the experimental results to account for slight misalignment of the imaging system.

Under the assumption that probe refraction can be neglected, we can assign an intensity value to each (x,y) point. Conversion of the measured phase shift to the nonlinear refractive index is possible because the interaction length through the target is known through a separate interferometry measurement [1]. The ionization yield as a function of peak intensity extracted from our measurements is shown in Fig. 2, as is a fit of the data to $\sigma I^m$ (dashed lines).

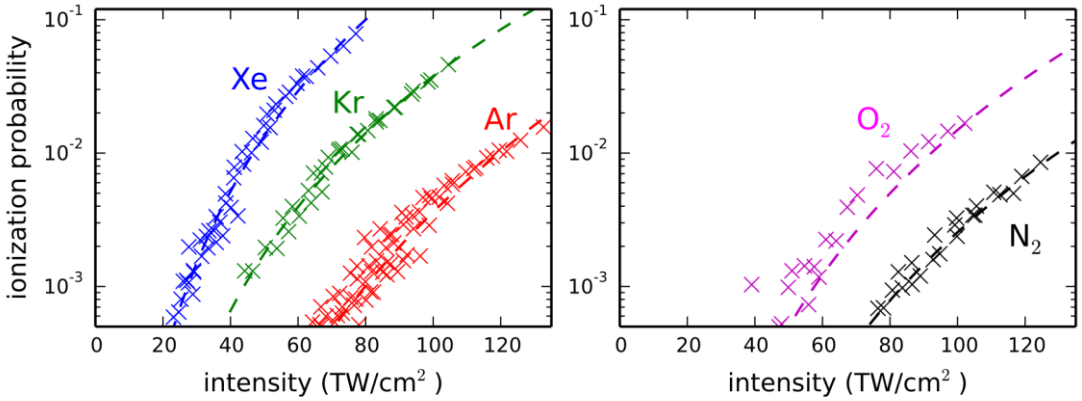


Fig. 2. Ionization probability vs. intensity in Ar, Kr, Xe, $N_2$, and $O_2$. Ionization yield (x) as a function of peak intensity for (a) Ar (red), Kr (green), Xe (blue); (b) $N_2$ (black) and $O_2$ (magenta).

The phase shift during the pump pulse has contributions from both the Kerr effect and ionization. The time dependent phase shift is shown in Fig. 3 for Ar. The total phase shift is consistent with the simple addition of these two contributions. Also shown in the figures are fits of the data to $\Delta n(t) = \Delta n_K e^{-t^2/\tau^2} + \Delta n_p \,\mathrm{erf}[t/(\tau/m^{1/2})]$. It is seen that the approximately linear dependence of $\Delta n_K$ on intensity measured at low intensities [2] continues at high intensity. The Kerr component may actually be slightly higher than the low intensity slope. Further investigations of this effect are underway.

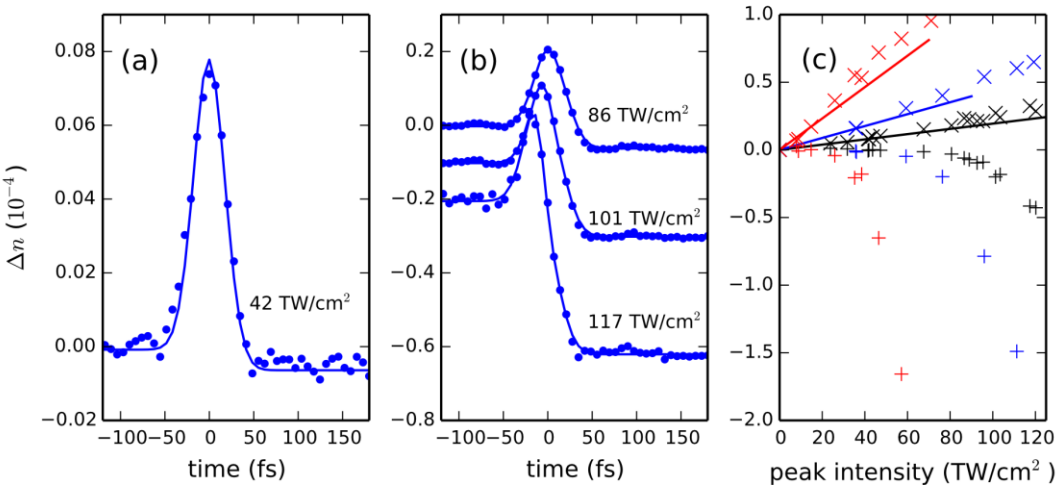


Fig. 3. Response during pump pulse. (a) The nonlinear refractive index as a function of time for peak intensity of 42 TW/cm$^2$, below the threshold for ionization, and a fit to a Gaussian with FWHM 42 fs. (b) Nonlinear refractive index as a function of time and fits to the standard model (Kerr effect plus ionization) for Ar. The curves have been offset vertically for clarity. (c) Plot of Kerr index change (×) and plasma index change (+) from fits as a function of intensity in Ar, Kr, and Xe.


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