High Field Optical Nonlinearity and the Kramers-Kronig Relations

J. K. Wahlstrand, Y.-H. Cheng, and H. M. Milchberg

Institute for Research in Electronics and Applied Physics, University of Maryland, College Park, Maryland, 20742, USA

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The nonlinear optical response to high fields is absolutely measured for the noble gas atoms He, Ne, Ar, Kr, and Xe. We find that the response is quadratic in the laser field magnitude up to the ionization threshold of each gas. Its size and quadratic dependence are well predicted by a Kramers-Kronig analysis employing known ionization probabilities, and the results are consistent with calculations using the time-dependent Schrödinger equation.

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The high field nonlinear optical response of gases is important to a wide range of variety of phenomena under intensive investigation for applications, including attosecond physics [1], femtosecond filamentation [2], pulse compression [3], and high power atmospheric propagation [4]. Most of these applications involve exposing atoms to laser fields up to and beyond their ionization thresholds, so an important question has arisen regarding the nonlinear response of atoms at these high fields, where perturbation theory is expected to fail. Once ionization occurs, the plasma response of the free electrons typically dominates that of the bound electrons. However, much of the dynamics of self-phase modulation and self-focusing takes place just short of ionization and so the bound electron response in that regime is of high interest. Behavior in this regime meets its most sensitive test in femtosecond laser pulse filamentation, where substantial temporal slices of a pulse envelope experience propagation just below and just above the ionization threshold.

In this Letter, we present experiments showing that for the noble gases He, Ne, Ar, Kr, and Xe, the nonlinear response is quadratic in the laser field strength (linear in the intensity) up to the rapid onset point at which free electrons measurably perturb the refractive index, which we deem the ionization threshold. We show that by employing known gas ionization rates in the generalized Kramers-Kronig (KK) relations, we recover a real nonlinear response which scales linearly with intensity, in good agreement with our experiments. The results are consistent with calculations using the time-dependent Schrödinger equation.

Our experiment directly measures the nonlinear response of thin gas samples using single-shot supercontinuum spectral interferometry, which allows extraction of the time- and 1D space-dependent nonlinear phase shift $\Delta\Phi(x, t)$ imposed on a probe pulse by a pump-induced refractive index change, where x is a spatial coordinate across the pump spot. The experimental setup and data extraction have been described recently [5]. The laser is a 1 kHz repetition rate Ti:sapphire amplifier producing 40 fs, 3.5 mJ pulses. Supercontinuum with a bandwidth of 120 nm is generated in an SF_6 filamentation cell [6] and split into two replica pulses separated by 2 ps by a Michelson interferometer. The pump, probe, and reference pulses are collinearly combined using a dichroic mirror and focused into a thin gas flow ($L_{\text{eff}} \sim 200$–400 μm, depending on the gas used) in a laser-drilled tube in a vacuum chamber pumped by a roots blower. An auxiliary spatial interferometry beam [7] allowed extraction of the absolute linear phase shift of the Ar stationary gas flow; all other gases were referenced to Ar, providing absolute calibration of $\Delta\Phi(x, t)$. The supercontinuum probe and reference are imaged from the gas onto a CCD at the image plane of a spectrometer and interfere in the frequency domain, allowing extraction of $\Delta\Phi(x, t)$ by Fourier techniques [8]. We average 1000 interferograms before performing the extraction, allowing measurement of phase shifts as small as 5×10^{-3} rad. The nonlinear refractive index shift is $\Delta n(x, t) = (k_0 L_{\text{eff}})^{-1} \Delta\Phi(x, t)$. The absolute index shift is calibrated by a measurement in a backfilled vacuum chamber at low pressure, as in [7]. Figure 1 (left panel) shows two $\Delta\Phi(x, t)$ traces for neon, one at a peak intensity below where plasma is generated and one just above. We find for all gases measured that, within experimental error, Δn is linear in the intensity envelope up to the point at which plasma contributes to the phase shift, at which point a negative contribution to the phase envelope appears at the peak of the pulse and later in time. Below the ionization threshold, we write $\Delta n(x, t) = 2 n_2 I(x, t)$, where $I(x, t)$ is determined directly from the pump pulse energy and from $I(x, t) \propto \Delta\Phi(x, t)$ at low intensity in Ar. The factor of 2 recognizes that Δn describes cross-phase modulation and n_2 is the nonlinear self-refractive index.

It is well known that the real and imaginary parts of the linear refractive index $n(\omega)$ are related through causality (equivalent to analyticity of $n(\omega)$ in the upper half plane) by the Kramers-Kronig relations [9]. It is perhaps less appreciated that these relations can also apply in the nonlinear case, where a strong applied or pump field $E(\omega')$ can be considered a parameter of the pump field plus matter system experienced by a weak probe of frequency Ω. The application of KK relations to nonlinear interactions has

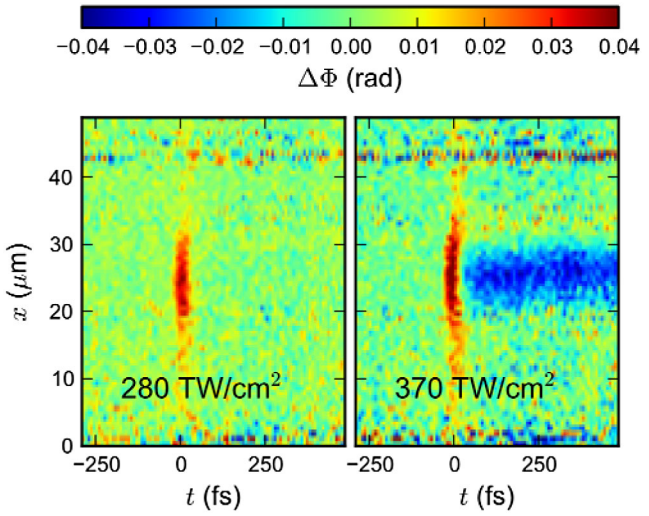

FIG. 1 (color online). Probe phase shift $\Delta\Phi(x, t)$ measured in neon using single-shot supercontinuum spectral interferometry, for an intensity below (left) and above (right) the onset of ionization.

been reviewed by Hutchings, Sheik-Bahae *et al.* [10], and has been used to calculate the real nonlinear index of refraction (n_2) of semiconductors from two-photon absorption, Raman transitions and the ac Stark effect [11]. In this analysis, they related the real and imaginary parts of the response quadratic in the field strength.

To apply the general KK relation to nonlinear absorption and the nonlinear refractive index [10], one considers the linear response to a weak probe field at frequency Ω of the system consisting of the matter and a strong optical pump field $E(\omega')$. The real refractive index change Δn for a probe field at frequency ω is then

$$\Delta n(\omega, E(\omega')) = \frac{c}{\pi} P \int_0^\infty \frac{\Delta\alpha(\Omega, E(\omega'))}{\Omega^2 - \omega^2} d\Omega, \qquad (1)$$

where P denotes principal value. Here, $\Delta\alpha(\Omega, E(\omega'))$ is the change in absorption coefficient of a probe beam at frequency Ω in the system composed of the matter and a pump field $E(\omega')$. Below we are interested only in the index change for nearly degenerate fields, so we set $\omega' = \omega$ after performing the integration [10]. For the Kerr and ionization processes discussed in this Letter, where the response time is essentially instantaneous and thus only the pump field magnitude is important, we replace $E(\omega')$ in Eq. (1) with intensity $I(\omega')(\propto |E(\omega')|^2)$ and write $\Delta\alpha(\Omega, I(\omega')) = \Delta\alpha(\Omega, \omega', I)$.

In recent papers, Brée *et al.* [12–14] applied this procedure to high order processes in gases and transparent solids: they derived the real part of the kth order response ($n_{2k} I^k$) from the kth order absorption process, which they identified as kth order multiphoton ionization (MPI). The corresponding absorption coefficient is proportional to $I^{-1} w_{k+1}$, where $w_k = \sigma_k I^k$ is the kth order MPI rate. They found that the nonlinear index shift for argon obtained from the series $\Delta n = \sum n_{2j} I^j$ saturates and turns negative in the intensity range $I \sim 40$–50 TW/cm^2, well below the threshold for ionization, which is near ~ 80 TW/cm^2. At very low intensity, their method gives reasonable agreement for the $k = 1$ case (n_2) with prior experimental results for the noble gases [12,13].

Equation (1) formally requires knowledge of the absorption spectrum as a function of the frequencies Ω and ω', which available theories based on monochromatic optical fields cannot provide. Brée *et al.* [12–14] extended an "average frequency" approximation used by Sheik-Bahae *et al.* for two-photon absorption [11] to arbitrary order K-photon processes, using $\Delta\alpha(\omega_1, \omega_2, \ldots, \omega_K) \approx \Delta\alpha((\omega_1 + \cdots + \omega_K)/K)$. For absorption of K pump photons at frequency ω' and one probe photon of frequency Ω, the change in absorption was thus written as

$$\Delta\alpha(\Omega, \omega', I) \approx \Delta\alpha(\omega_{\mathrm{avg},K}, I), \qquad (2)$$

where $\omega_{\mathrm{avg},K} = (K\omega' + \Omega)/(K + 1)$, which assumes excitation with a single average frequency. We also adopt this approach, but examine its accuracy below.

In any consideration of high field ionization as an absorption mechanism to be used in Eq. (1), one must be aware of the transition between perturbative (MPI) and nonperturbative (tunneling) ionization regimes. A measure of this transition is the dimensionless Keldysh parameter $\gamma = (2\chi_p)^{1/2}\omega/E_0$, where χ_p is the ionization potential of a gas atom, E_0 is the peak laser field, and atomic units are used. While one is safely in the MPI limit for $\gamma \gg 1$ and in the tunneling limit for $\gamma \ll 1$, it is typically qualitatively argued that $\gamma < 1$ is sufficient to be in the tunneling limit and $\gamma > 1$ to be in the MPI limit [12]. As we show here, when considering the KK relations, one must be considerably more careful.

In their calculation, Brée *et al.* use expressions for the k-photon ionization rate w_k derived from the $\gamma \to \infty$ or MPI limit of a recently derived ionization rate R [15] based on the theory of Perelomov-Popov-Terent'ev [16]. The expression R (Eq. 6 of [15]) is valid at arbitrary values of γ. It handles the MPI and tunneling limits, and provides an accurate interpolation for γ lying in between. It assumes a single active electron and nonrelativistic intensities. In Fig. 2(a), we plot R, R_T, and $w = \sum_{k=11}^{50} w_k$ for atomic hydrogen ($\chi_p = 13.6$ eV) for $\hbar\omega = 1.55$ eV ($\lambda = 800$ nm) as a function of intensity, where R_T is the tunneling rate [17] (equal to the $\gamma \ll 1$ limit of R) and the $k > 9$ terms in w represent above threshold ionization. At low intensities $R \propto I^M$, where $M(= 9$ for H) is the minimum number of photons for ionization, and at higher intensities R rolls off, asymptotically approaching R_T. Above ~ 70 TW/cm^2 ($\gamma = 1.3$), w diverges significantly. However, even as low as 6 TW/cm^2 ($\gamma = 4.4$), R and w differ by a factor of ~ 100. Similar plots for Ar show that the terms containing σ_k in w are inadequate to describe ionization in the

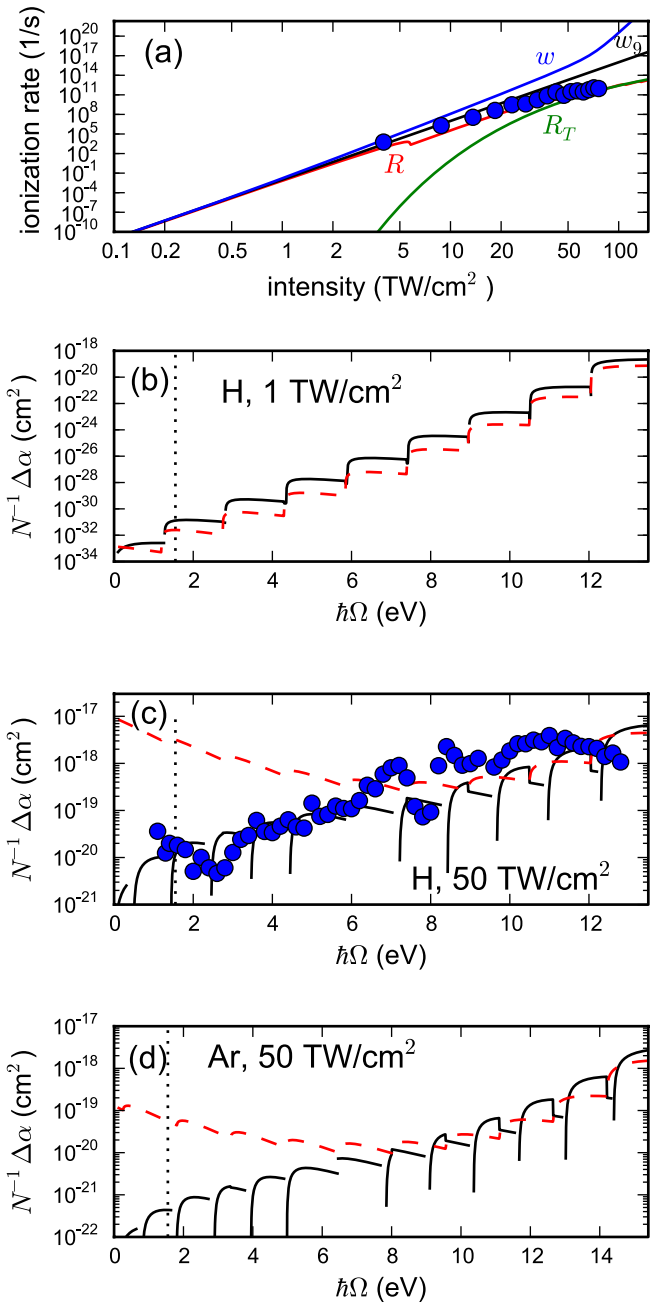

FIG. 2 (color online). (a) Ionization rates w, w_9, R, and R_T calculated using the models in [12,15,17] for 800 nm light in H. Here w_k is the kth order MPI rate applicable for $\gamma \gg 1$ (shown for $k = 9$), w is the sum of w_k from $k = 2$ to 50, R_T is the rate in the tunneling limit $\gamma \ll 1$ [17], and R is the rate from [15] applicable from the MPI through tunneling regimes. The blue dots are for a TDSE calculation. (b) Calculation of the absorption cross section $N^{-1}\Delta\alpha$ in Hydrogen using R (solid black line) and the $\Delta\alpha$ expression from [12] (dashed red line) for pump intensity 1 TW/cm^2 and $\hbar\omega = 1.55$ eV. (c) Curves of (b) for pump intensity 50 TW/cm^2. The blue dots show the probe absorption cross section derived from the nondegenerate ionization rate determined by a TDSE calculation [19]. (d) Same as (b) and (c) but in Ar at 50 TW/cm^2. Vertical dotted lines in (b), (c), and (d) show where $\Omega = \omega$. For $\Omega < \omega$ the KK integrand in Eq. (1) is negative.

>40 TW/cm^2 region ($\gamma < 1.7$) where the novel behavior of Δn is claimed by Brée *et al.* [18].

The consequences of the divergence in the MPI ionization rate for small γ are apparent in plots, shown in Fig. 2, of the absorption coefficient per unit gas density $N^{-1}\Delta\alpha$ (absorption cross section) as a function of probe frequency Ω, where $\Delta\alpha$ appears in the numerator of the KK integral [Eq. (1)] and sensitively determines the calculated real nonlinear index shift. The steps in the coefficient are the onset of new multiphoton resonances as probe frequency is increased. When the tunneling regime is approached, the multiphoton thresholds become less defined. Figures 2(b) (for pump intensity 1 TW/cm^2) and 2(c) (for pump intensity 50 TW/cm^2) compare the coefficients $N^{-1}\Delta\alpha_R$ derived from the full rate R (black solid line) [18] and $N^{-1}\Delta\alpha_{\mathrm{MPI}}$ from the MPI-based rate w (red dashed line) [12]. Here, the average frequency is used in both R and w, where in R we use Eq. (2) with $K(\Omega) = [(\chi_p\text{-}\hbar\Omega)/(\hbar\omega)] + 1$ for $\hbar\Omega < \chi_p$ and otherwise $K = 1$, where the square brackets denote the integer part. This is equivalent to assuming that at a given probe frequency, the lowest-order nonlinear ionization process dominates.

At sufficiently high values of Ω and low pump intensity (larger values of γ), where the MPI model is accurate, it is seen in Figs. 2(b) and 2(c) that $\Delta\alpha_{\mathrm{MPI}}$ and $\Delta\alpha_R$ converge. However, their divergence greatly increases with increasing intensity over a wide range of Ω as seen in the change from Fig. 2(b) to Fig. 2(c). In Eq. (1), the region $\Omega < \omega$ contributes negatively to Δn, and at increasing intensity the MPI model strongly overestimates $\Delta\alpha$ in this frequency range, as seen in Fig. 2(c) and in Fig. 2(d), for Argon at 50 TW/cm^2. This is the source of the saturation and inversion calculated in [12,13]. A detailed analysis describing this saturation and inversion can be found in the Supplemental Material [18].

To study the accuracy of the average frequency assumption [Eq. (2)], we calculated the nondegenerate probe absorption $\Delta\alpha(\Omega, \omega', I)$ using a time-dependent Schrödinger equation (TDSE) simulation [18], using the publicly available computer code QPROP [19]. For a single frequency, it has been shown [15] to agree with the ionization rate R, as also seen in Fig. 2(a). Note that numerical TDSE studies have been used to directly calculate the nonlinear response (without a KK calculation) [20,21]. The nonlinear response has also been calculated using simplified models [22,23]. Here, we calculate the ionization rate $S_{\mathrm{TDSE}}(\omega, I_e, \Omega, I_p)$ of hydrogen exposed to a 2-color laser field: a strong pump I_e (50 TW/cm^2, $\hbar\omega = 1.55$ eV) plus a weak probe I_p (0.1 TW/cm^2, with variable $\hbar\Omega$). Using S_{TDSE}, in Fig. 2(c) we plot the absorption cross section $N^{-1}\Delta\alpha_{\mathrm{TDSE}} = \hbar\Omega(\partial S_{\mathrm{TDSE}}/\partial I_p)$ as a function of $\hbar\Omega$ (blue circles). It agrees reasonably well over the full range of Ω with $\Delta\alpha_R(\omega_{\mathrm{avg}}, I)$, providing some confidence

in the average frequency assumption [Eq. (2)], at least as applied to hydrogen. Note, however, that $\Delta\alpha_{\mathrm{TDSE}}$ deviates strongly from $\Delta\alpha_{\mathrm{MPI}}$.

Before we proceed to compute Δn from Eq. (1) using the various models for ionization-based $\Delta\alpha$, it is important to verify whether in fact ionization *is* the dominant absorption mechanism. Intuitively, one would expect that any intermediate excited states, which are strongly Stark shifted in the intense pump field, would be quickly depleted by ionization. TDSE calculations confirm that for the full probe frequency range, a negligible population is left in excited states; the dominant absorption channel *is* ionization. Only for narrow regions of probe energy $\hbar\Omega$, as seen near 8 eV in Fig. 2(c), is there apparent resonant structure. However, this reduced absorption structure, whose location in $\hbar\Omega$ shifts with pump intensity, is related to interferences of the outgoing electron wave packet, and there is no residual excited state population associated with it. The structure has little effect on the integral in Eq. (1).

We now compute Δn for He, Ne, Ar, Kr, and Xe directly from Eq. (1). We integrate Eq. (1) using $\Delta\alpha_R(\omega_{\mathrm{avg}}, I)$ from $\hbar\Omega = 0$ up to $2\chi_p - \hbar\omega$, then switch to $\Delta\alpha_{\mathrm{MPI}}(\omega_{\mathrm{avg}}, I)$, which allows removal of linear contributions from one-photon absorption for $\hbar\omega_{\mathrm{avg}} > \chi_p$. The result is shown in Fig. 3, where we have also overlaid our experimental values of Δn measured up to the ionization threshold in each gas [24]. The experimental results clearly confirm $\Delta n \propto I$ up to the ionization threshold, a remarkable result given that perturbation theory is hardly applicable at these intensity levels. The experimentally measured values of $n_2 = \Delta n/2I$ are in good agreement with previous estimates using harmonic generation [25], and are also close to the KK prediction in all cases. Note that no adjustable parameters were used in the experimental analysis or in the KK calculation. We see that using, in the KK integral, an ionization rate applicable from the MPI through tunneling regimes confirms the linear dependence of Δn on intensity. For comparison, we overlay the Brée *et al.* results [12]. This clearly demonstrates the adverse consequences of using perturbation theory outside its regime of applicability.

Overall, we find the agreement between our measured Kerr coefficients n_2 and the KK calculation remarkable given the idealizations present in the model for R. It is also interesting that the unsaturated intensity dependence predicted by KK analysis extends past our measured ionization thresholds. An intriguing interpretation of this result is that atoms probabilistically surviving ionization will continue to respond without saturation. Whether or not this is a real effect is a question best settled by experiment.

In conclusion, we show that the Kramers-Kronig relations can be successfully used to predict the nonlinear modifications to the refractive index of gases induced by an intense laser pulse. However, comparison with previous studies clearly highlights that correct results are only

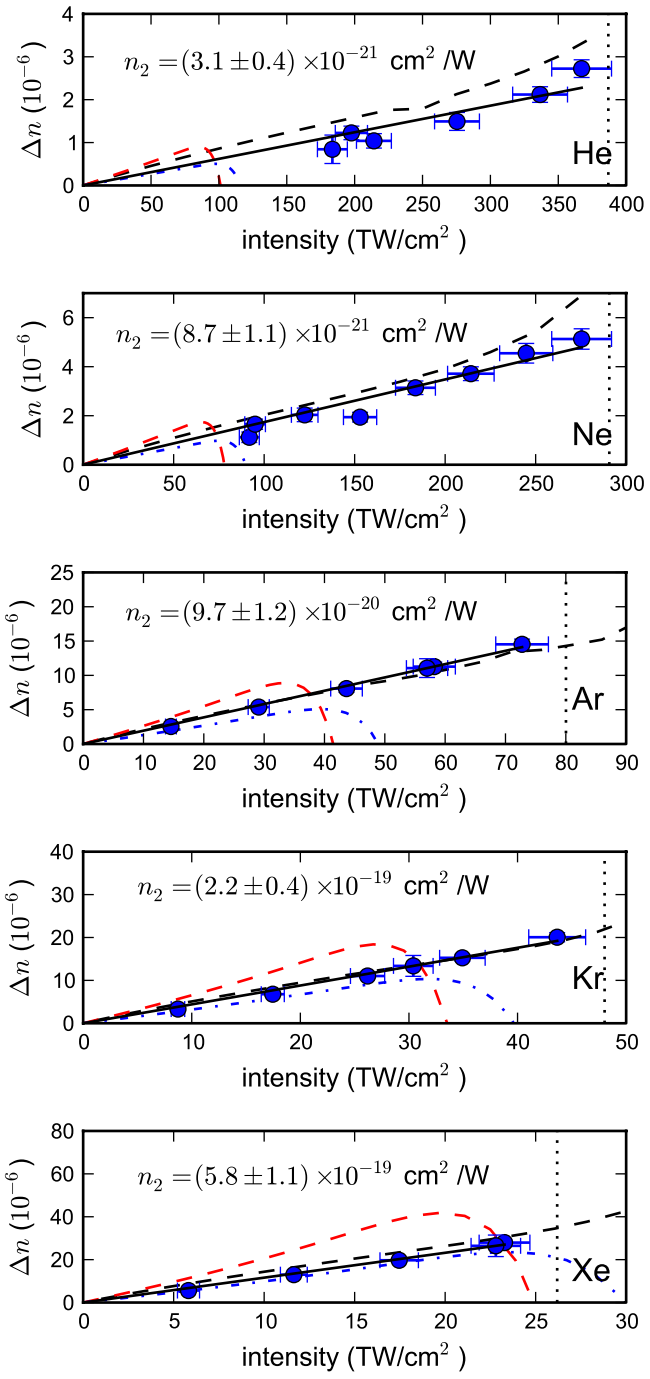

FIG. 3 (color online). The nonlinear refractive index Δn of the noble gases experienced by a weak probe pulse as a function of pump laser peak intensity. Blue dots show experimental data points, and the solid line shows a linear fit of the data to $2n_2I$. Vertical dotted lines show, for each gas, the measured ionization threshold. Our KK calculation, using the ionization rate R, is shown as a dashed black line. The red dashed line is the result from Brée *et al.* [12], where we have calculated Δn for a weak probe by multiplying their self-refractive index coefficients n_{2k} by $k + 1$ [18]. The blue dash-dotted line is the self-refractive index plotted in Brée *et al.* Horizontal error bars denote uncertainty in the intensity calibration and vertical error bars denote uncertainty in the phase extraction.

obtained by abandoning the standard perturbative approach adopted in nonlinear optics and including the full transition from multiphoton to tunneling ionization. A proper application of Kramers-Kronig analysis predicts a linear dependence of Δn on laser intensity up to the ionization threshold and gives results for n_2 in good agreement with experiments for a wide range of tested gases.

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