Measurement of pressure dependent nonlinear refractive index of inert gases

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Abstract: The propagation of high intensity laser beams is excessively affected by optical nonlinear effects, thereby the knowledge of the nonlinear refractive indices of the beam guiding media is indispensable in the design of laser systems and experiments. Apart from undesired self-focusing, several areas of modern laser spectroscopy can utilize optical nonlinearity, from LiDAR measurements to filamentation. In this paper we report on a direct measurement of pressure dependent nonlinear refractive index of Ar, N_2, Ne, Xe, and air between 0.05 mbar and 1 bar, based on the powerful technique called spectrally and spatially resolved interferometry. In this way the total value of nonlinear refractive index is measured, that is the sum of all elementary phenomena contributing to the intensity dependent refractivity of the gases.

OCIS codes: (070.4340) Nonlinear optical signal processing; (120.5050) Phase measurement; (190.3270) Kerr effect; (190.4400) Nonlinear optics, materials; (320.7110) Ultrafast nonlinear optics.

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1. Introduction

In experiments and laser systems, where high intensity laser beams interact with or propagate in gaseous media, the precise knowledge of the nonlinear refractive index (n_2) is essential. Namely, beam delivery of high power laser systems has to be designed so that the n_2 related effects as self-phase modulation and self-focusing should be avoided [1]. Moreover, many applications of modern laser technology can benefit from them, like atmospheric LiDAR experiments [2,3], white continuum generation [4,5], and more importantly filamentation, which can even lead to pulse compression [6–13].

It is well known that the smallest order Kerr-type nonlinearity is described as a shift in refractive index, which is proportional to the intensity. The scaling factor, called nonlinear refractive index, is caused by several elementary phenomena. Besides the instantaneous electronic contribution, molecular gases with anisotropic polarizability have additional rotational and vibrational contribution to the nonlinear refractive index [14–16]. Most recently, a theoretical calculation on the nonlinear refractive indices of inert gases has been presented by Breé et al. [17]. Their model uses Keldysh theory with Kramers-Kronig transformation to estimate the wavelength dependence of n_2, while the pressure of the gases is not integrated into the calculations.

The experimental studies of n_2 started in 70's when nanosecond pulse technology started allowing for the generation of high intensity pulses. Along with the development of laser technology, the Z-scan method proved to be the breakthrough in the early 90's [18]. That simple and powerful technique provided the experimental base for the vast majority of nonlinear measurements. Considering the technical difficulties and the necessary strong focusing, almost exclusively solids and liquids were the objects of Z-scan examinations.

To measure n_2 in gases, other techniques had to be developed, which utilized electric-field induced second harmonic generation [19], third harmonic generation [20], spectral distortion upon propagation [16,21], self-focusing [22,23], post-pulse molecular alignment [24], and most frequently, spectral interferometry [25–32]. Some of them are dealing with the time dependence of n_2 [25,27,28,31,32] and refractive index bursts [14]. However, when it is excited by ultrashort laser pulses, latter phenomena can be treated by collisionless gas model, that is, molecule structure can be considered to be frozen. Since in the engineering of high intensity laser systems [33,34] and the aforementioned bandwidth broadening processes basically the sign and gross value of n_2 are of the highest interest, present work is regarding n_2 as only the compound of the aforementioned nonlinear effects, which retroact to the exciting pulses. Although the net nonlinear effect depends on not only the intensity but also pressure, it is worth mentioning that apart from one measurement made in the pressure range of 0.2 and 3 bars for a N_2, O_2 and air [16], there is no comprehensive data set for the pressure dependence of n_2 over more orders of magnitude.

Spectral interferometry, one of the most common measurement methods of n_2 in gases, is based on the combination of a two-beam interferometer and a (one dimensional) spectrograph. The beams from the reference arm and the sample arm are collinear, while the pulses are slightly delayed. The spectrally resolved interference pattern is ultimately characteristic to the object in the sample arm. In a so-called spectrally and spatially resolved interferometer (SSRI) [35] the phase fronts of the sample and reference pulses are not parallel anymore, while the sample and reference pulses are temporally overlapped. This method, developed for accurate measurement of linear dispersion of various materials from chirped mirrors through gases to complicated systems [36–38], offers considerably higher precision than SI [39]. Recently, SSRI was also applied to study nonlinear phenomena [40]. SSRI may be also an appropriate tool for measuring n_2 in gases [41], as the high sensitivity of SSRI allows to reduce further the minimum pulse intensity propagating through the sample gas, so that no spatial change of beam profile [42] could be expected.

In this paper we report on measurement of pressure dependent nonlinear refractive index of inert gases as Ar, Ne, Xe as well as air and N_2 with the novel use of spectrally and spatially

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resolved interferometry. The total value of n_2 experienced by a laser pulse has been determined independently of their physical origin.

2. Description of spectrally and spatially resolved interferometry

The object, positioned into the sample arm of an SSRI, shifts the spectral phase of the sample pulse. The reference pulse propagates undisturbed in the other arm of the two beam interferometer. Since phase fronts of the sample and reference pulses are tilted with an angle γ, so that the temporally overlapped pulses leaving the interferometer form fringes at the input slit of an imaging spectrograph. The interferograms captured by the CCD camera of the spectrograph are hence spectrally and spatially resolved along the dispersion plane of the spectrograph (frequency axis, ω) and the slit of the spectrograph (spatial axis, y), respectively. The intensity distribution of the interference pattern these interferograms can be written as

$$I(y,\omega) = I_R(y,\omega) + I_S(y,\omega) + 2\sqrt{I_R(y,\omega)I_S(y,\omega)}\cos\left[\frac{\omega}{c}\cdot 2\gamma(y-y_0) + \Delta\phi(y,\omega)\right], \quad (1)$$

where c is the velocity of light, and $I_R(y,\omega)$ and $I_S(y,\omega)$ are the intensity distributions in the reference and sample arm, respectively. The first term inside the argument of the cosine function describes the spatial modulation of the interference pattern. The other one is the difference between the $\varphi_S(y,\omega)$ and $\varphi_R(y,\omega)$ phase shifts of the arms, which includes the phase shift introduced by the investigated object in the sample arm. This term determines the shape of the SSRI fringes, from which eventually the spectral phase shift of the sample can be obtained.

The idea of the measurement of nonlinear refractive index of a gas sample is that the pulse intensity in the sample arm shall be considerably higher than in the reference arm. The intensity even in the sample arm is assumed so weak that its spatial distribution is kept along the propagation. Finally, it can be also ensured that the reference and the sample pulses passing through the same amount of transparent materials (like beam splitters) at the same intensity so that any further nonlinear phase distortion arising from these parts could be neglected. Under these circumstances, the spectral phase shift of the sample and reference pulses can be calculated as

$$\phi_S(y,\omega) = \frac{\omega}{c}\left(\left(n^{Med}(\omega) + n_2^{Med}\int I_S(y,\omega)d\omega\right)\cdot l_{S,Med} + \left(n^{Obj}(\omega) + n_2^{Obj}\int I_S(y,\omega)d\omega\right)\cdot l_{Obj}\right), \quad (2)$$

and

$$\phi_R(y,\omega) = \frac{\omega}{c}\left(\left(n^{Med}(\omega) + n_2^{Med}\int I_R(y,\omega)d\omega\right)\cdot l_{R,Med}\right), \quad (3)$$

where l_{Obj}, $l_{S,Med}$ and $l_{R,Med}$ is the geometrical length of the gas object, the rest of the sample arm and the reference arm, respectively. n^{Obj} and n^{Med} are refractive indices of the sample object and the medium outside the object, which the pulses propagate in (usually ambient air); n_2^{Obj} and n_2^{Med} are nonlinear refractive indices, likewise before.

Evaluation of the captured interferograms provides the spatially dependent relative spectral phase shift $\Delta\varphi(y,\omega)$. From the measurement of the spatial beam distribution, energy and duration of the sample and reference pulses, and calculating the linear refractive indices from [37], eventually the nonlinear refractive index of the object can be deducted.

3. Model calculations

To optimize the experimental conditions and evaluation, theoretical calculations have been carried out. Let us assume that the intensity of the Gaussian sample pulse can be varied over magnitudes while the intensity of reference pulse is kept negligible compared to the sample pulses. In this approximation the nonlinear refractive index will basically contribute to the spectral phase shift of the sample pulses only. The sample medium is air with l_{Obj}, = 10 m and $l_{S,Med}$ = 0 m, while the sample and reference pulses are with a bandwidth of 50 nm, central frequency of 800 nm and a Gaussian beam waist of 0.5 cm. As one can see from the simulated interferograms in Fig. 1, there is no visible trace of nonlinear phase distortion at 25 GW/cm^2,

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while the fringes are spectacularly distorted at 2.5 TW/cm^2. It is worth mentioning, however, that the sensitivity of the SSRI method makes possible to detect nonlinear spectral phase shift at sample pulse intensity considerably lower than 25 GW/cm^2.

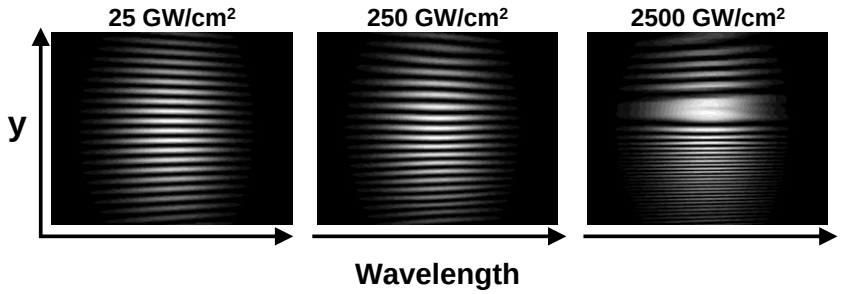

Fig. 1. Spectrally and spatially resolved interference fringes calculated for various intensity of transform-limited pulses propagating 10 m in ambient air.

Another observation helps in the evaluation of the fringes. It was demonstrated that the more chirped the input pulse the more similar is the spectral evolution of the nonlinear phase shift to the shape of the spectral intensity [43]. Hence, we optimized the pulse duration by model calculations to find the minimum chirp level when the spectral phase curve and the spectral intensity are overlapping satisfactorily. We have found that stretched pulse durations at least 8 times longer than the transform limited length satisfy this criterion. Figure 2 shows the evolution of similarity from transform limited shape to even more chirped pulses compared to the spectral intensity (blue curve).

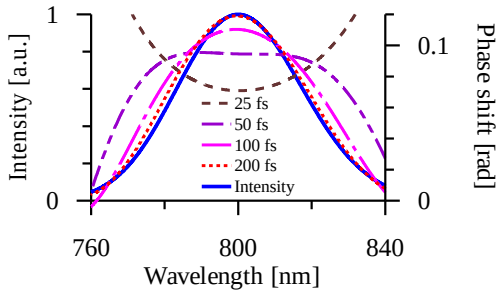

Fig. 2. Calculated spectral shape of nonlinear phase shift at different stretched pulse durations of a transform limited 25 fs pulse.

4 Experimental

In the experiment 800 nm pulses from the preamplifier of a CPA laser system were used. The Mach-Zehnder interferometer of the SSRI setup was basically built into a vacuum tube (Fig. 3).

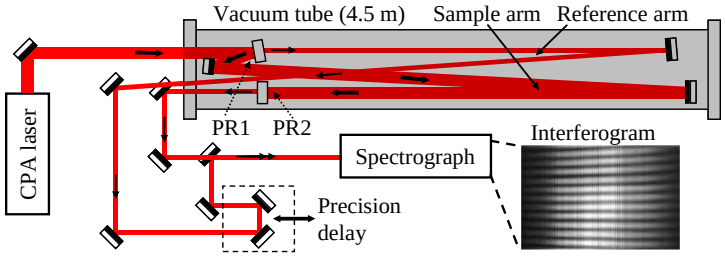

Fig. 3. Experimental setup. The intensity ratio of the sample and reference arm is 10:1, while their intensity is equal at the input and output windows of the tube.

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In accordance with the modeling described above, the incoming pulses with a bandwidth of 40 nm were left stretched to a duration of 200 fs. An experimental proof of the similarity between the spectrum and the spectral phase shift at this pulse duration is shown in Fig. 4.

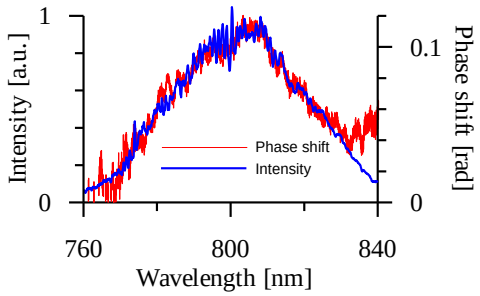

Fig. 4. Experimental verification of the similarity between the spectral shape of nonlinear phase shift of the stretched pulses and the spectral intensity of the pulses (c.f. Fig. 2).

The energy of the pulses was kept below 1 mJ, close to the limit of noticeable effects of self-phase modulation on the spectra and self-focusing on the beam size when the gases were at ambient pressure. The collimated, spatially Supergaussian beam with a diameter of 2 mm was split unequally inside the chamber by a partial reflector (PR1), so that an intensity ratio of 10:1 is formed between the sample and reference arm. Both the reference and sample pulses propagated approximately l_{tube} = 9 m in the tube and left it through a fused silica window. Just before leaving the tube, however, the intensity of the sample pulses was reduced by a partial reflector PR2 to the intensity level of the reference pulses. The partial reflectors PR1 and PR2 introduce equal phase shift in the interferometer arms, hence in this arrangement both the sample and the reference pulses had the same intensity upon propagating through the same amount of dispersive material of PR2 and PR1, respectively, and they are chirped by the same material. Thus, the spectral phase difference between the sample and reference beams leaving the tube can be eventually accounted for the nonlinear phase shift in the sample gas due to the unequal intensities in the two arms. When the adequate quantities are substituted into Eqs. (2-3), the phase difference is written in the form of

$$\Delta\phi(y,\omega)=\frac{\omega}{c}\left(n_2\left(\int I_S(y,\omega)d\omega\cdot l_{tube}-\int I_R(y,\omega)d\omega\cdot(l_{tube}-l_{corr.})\right)-(n^{air}(\omega)-n(\omega))\cdot l_{corr.}\right),\quad(4)$$

where $l_{corr.}$ = 47 cm is the difference in propagation length in the vacuum chamber.

The sample and the appropriately delayed reference pulses were then overlapped and sent to the entrance slit of an imaging spectrograph. The captured interferograms are spectrally and spatially resolved, with ranges of approximately 120 nm and 3 mm, respectively. The measurements were carried out at an ambient temperature of 20°C and within the pressure range from 0.05 mbar to 1020 mbar. We have divided this range to 5 logarithmically equidistant values over each order of magnitude, and carried out two sets of measurements: the one upon increasing and the next one upon decreasing the pressure. An oil free vacuum pump was used to evacuate the tube, while the pressure was measured by two of capacitance gauges independently.

At a certain pressure nine interferograms have been captured. The phase information of every interferogram is retrieved by a special scanning cosine fit algorithm called phase mapping, which is detailed in [39]. Having the phase maps processed, the spectral phase shift was obtained from the spatial and spectral distribution of the incoming pulse (similar to Fig. 4.). The nonlinear refractive index was then determined from Eq. (4), where n^{air} was calculated from [37].

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5. Results

A typical set of measurements showing the pressure dependent n_2 of argon, air and neon are displayed in Fig. 5 a-c. Similar curves have been measured for nitrogen and xenon, too. The dependence of the non-linear refractive indices on the pressure is linear, as it was expected. The measured n_2 values specific to pressure are listed in Table 1, along with the previously reported data.

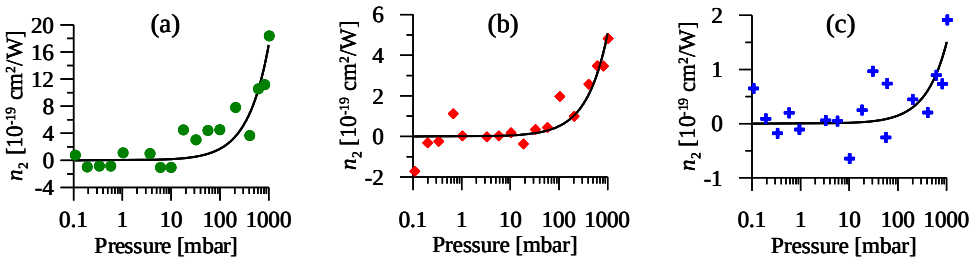

Fig. 5. Pressure dependence of n_2 for (a) argon, (b) air and (c) neon. Similar graphs have been obtained for xenon and nitrogen, too. Linear regressions are represented by black curves. Please mind the logarithmic scale.

The relative error of the present measurement seems higher than expected from our previous measurements on the linear refractive indices [37]. The possible reason is that the correspondence between the spectral phase shift and intensity spectrum might have been mediocre at some cases. However, the measured n_2 values at 1 bar pressure are in agreement with the measured and theoretically deducted values from earlier works.

It is worth noting that the n_2 values reported so far, including the present work, are ranging over one order of magnitude (Table 1). The reason behind the lowest values may be that those are corresponding to a single physical phenomenon like hiperpolarisability, while the works reporting larger values consider impact of several phenomena, or measure the result of contribution of all physical processes to the n_2 value as reported in this paper.

Table 1. Pressure-dependent nonlinear refractive indices for Ar, Ne, Xe, N_2 and air. For comparison, the previously reported theoretical [17] and experimental [16,19,24,32,44] n_2 values at 1000 mbar are also displayed.

	Argon	Air	Neon	Xenon	Nitrogen
Pressure dependent n_2 [× 10^{-19} cm^2/W at 1bar]	19.4 ± 1.9	5.7 ± 2.5	1.8 ± 1.5	84.3 ± 42.0	6.7 ± 2.0
Values from the literature at 1 bar [× 10^{-19} cm^2/W]	0.98[16], 7.96[17], 2.0[24], 7.3[32] 1.74[19, 44]	5.57[16], 2.4[24]	0.85[17] 0.14[19, 44]	54.8[17] 11.15[19, 44]	4.52[16], 2.2[24], 12.7[32] 1.36[19]

The feasibility of our experimental condition aiming at the use of weak short pulses to avoid nonlinear distortions was checked by deducting the B-integral from the measurements. Note, that the B values are directly derivable from interferograms as the total nonlinear phase shift. The highest B-integral of 0.121 was found at argon. Values for the other gases are 0.036 rad for air, 0.011 rad for neon, 0.053 rad for xenon and 0.042 rad for nitrogen. All of these results are obtained within the conditions of 1000 mbar and 293 K, except for xenon with a peak pressure of 100 mbar.

6. Summary

As a conclusion, we have measured the pressure dependent nonlinear refractive indices of inert gases with the use of spectrally and spatially resolved interferometry. In our measurement the total value of n_2 experienced by a laser pulse was determined, that is, the

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collective contribution of all possible laser induced elementary processes in the gases. In contrast to the widely used Z-scan method, the SSRI technique does not require extreme high pulse energies nor beam focusing, so it would be also applicable to *in situ* measurement of n_2 in experimental arrangements. In particular, we believe that the values of n_2 systematically determined with the same method for various inert gases at ambient pressure would be of high interest of the community dealing with high intensity pulses and nonlinear optical phenomena.

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