

# Investigation of two-photon-induced polarization spectroscopy of the $a-X(1,0)$ transition in molecular nitrogen at elevated pressures

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Two-photon-induced polarization spectroscopy of molecular nitrogen in the  $a^1\Pi_g(v' = 1) \leftarrow X^1\Sigma_g^+(v'' = 0)$  system near 283 nm was performed, and its signal dependence investigated over the pressure range from 1.2 to 5 bars at 300 K. A significant increase of the signal intensity with pressure beyond the expected square law for a two-photon process was observed for pure nitrogen. Similar behavior was also found for a constant nitrogen partial pressure with increasing partial pressures of argon buffer gas. In both cases the spectral linewidth of the excited transitions increased dramatically with overall pressure. A possible explanation is given for the observed behavior in terms of contributions to the nonlinear susceptibility of the medium from the population of one-photon resonantly absorbing excited-state nitrogen and ground state  $N_2^+$  ions created in the multiphoton absorption process at the high laser intensities required. © 2000 Optical Society of America

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## 1. Introduction

Multiphoton excitation of atoms and molecules is a convenient technique to overcome several drawbacks encountered in linear, single-photon-allowed spectroscopies. One advantage is the accessibility of atomic and molecular energy levels that, because of symmetry constraints, cannot be probed by one-photon transitions.<sup>1</sup> A second aspect is the avoidance of interferences in the detected signal caused by scattering of laser radiation from foreign species and particulates, or from containment walls, given that excitation and detection wavelength are spectrally well separated. For the study of reacting flows such as combustion, many important species (H, O, CO, etc.) have their first accessible electronic transitions in the vacuum ultraviolet spectral region. This poses a twofold problem inasmuch as no adequate excitation sources may be at hand, and, furthermore,

the ambient environment usually absorbs strongly at the required optical frequencies. Thus, for many species of interest, there are no alternatives to multiphoton excitation schemes. Within the context of combustion diagnostics multiphoton excitation of atoms and molecules has been successfully applied in the past. By use of absorption or laser-induced fluorescence, spatially resolved relative concentrations of atomic hydrogen,<sup>2</sup> oxygen,<sup>3</sup> nitrogen,<sup>4</sup> and the molecular species  $NH_3$ ,<sup>5</sup>  $CO$ ,<sup>6,7</sup>  $NO$ ,<sup>8</sup> or  $N_2$  (Ref. 9) have been measured.

Multiphoton excitation can also be exploited in nonlinear coherent spectroscopic approaches such as degenerate four-wave mixing<sup>10</sup> or polarization spectroscopy (PS).<sup>11</sup> Today PS is an established tool for high-resolution spectroscopy,<sup>12-14</sup> combining experimental simplicity with species selectivity. The technique has recently gained renewed interest as a tool in combustion and plasma diagnostics for the detection of atoms,<sup>12</sup> radicals,<sup>14,15</sup> stable molecular species,<sup>16</sup> temperature measurements,<sup>17</sup> and in two-dimensional imaging.<sup>18</sup> Although much progress has been made in the understanding of the technique,<sup>19</sup> there is still no complete theory available that accounts for all the aspects of the technique that can be encountered in practice.

The situation is even worse for two-photon-induced polarization spectroscopy (TIPS) for which there are only a few theoretical investigations available to

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date. Important contributions to the development of TIPS have been made experimentally by Danzmann *et al.*<sup>20</sup> and theoretically by Seidel.<sup>21</sup> The latter showed that, owing to the short time scale at which the TIPS process takes place, the signal should be unaffected by molecular quenching collisions. This point has major implications for application of the technique to species concentration measurements in reactive environments where the local concentrations of foreign collider species are unknown or can vary in time, which provided the motivation for the present study. To our knowledge no systematic investigation of the pressure dependence of TIPS signals has been reported. Here we use the well-studied  $a^1\Pi_g(v' = 1) \leftarrow X^1\Sigma_g^+(v'' = 0)$  transition in molecular nitrogen<sup>22,23</sup> that occurs near 283 nm. We conducted signal intensity and linewidth measurements at pressures ranging between 1 and 5 bars by using mixtures that contain nitrogen or argon as a major collision partner. The results complement previous research in our group,<sup>24</sup> which demonstrated the feasibility of TIPS of  $N_2$  at atmospheric pressure. We hope to promote further interest for theoretical treatments of multiphoton polarization spectroscopy to realize its potential for use as a diagnostic tool in practical environments.

## 2. Experimental

Radiation around 283 nm was obtained by frequency doubling the output of a grating tuned dye laser that operates with Rhodamine 590 dye in methanol. The dye laser was pumped by the frequency-doubled output from a Nd:YAG laser. Approximately 10 mJ/pulse were obtained in the UV in a measured bandwidth of  $0.15 \pm 0.03 \text{ cm}^{-1}$  at 7-ns pulse length. We employed a standard PS setup in a forward propagating beam geometry<sup>24</sup> with a strong pump (pulse energies ranging from 0.5 to 4 mJ) and a weaker ( $\sim 20\text{-}\mu\text{J}$ ) probe beam. Each beam was focused by a quartz lens of 250-mm focal length, and both beams were crossed near the focal point in the sample cell at an angle of  $4.5^\circ$ . We oriented the polarization planes of the linearly polarized pump and probe beams at an angle of  $45^\circ$  with respect to each other by using a double Fresnel rhomb in the pump beam path.

The gas samples were contained in a specially designed high-pressure stainless-steel cell as shown in Fig. 1 and equipped with O-ring-sealed quartz windows (50-mm diameter, 20 mm thick) for entrance and exit of the laser beams. The cell consisted of three connected cubes, each having dimensions of  $100 \text{ mm} \times 100 \text{ mm} \times 100 \text{ mm}$ . The central cube was fitted with connections for gas supply and pressure measurements as well as with a window for the simultaneous observation of laser-induced fluorescence (LIF). The end section cubes provided the necessary space for installation of the Glan–Taylor polarizer and analyzer prisms (Halle) in the probe beam path. The extinction ratio at the laser energies used in the probe beam was measured to be better than  $10^{-4}$  with polarizer and analyzer crossed. The polarizers

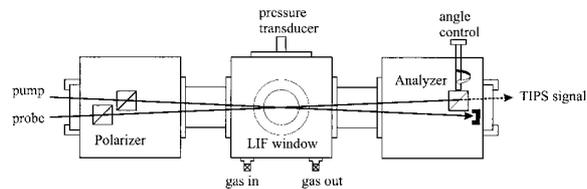


Fig. 1. Schematic drawing of the high-pressure stainless-steel gas cell used for the TIPS experiments. The polarization optics in the probe beam path are mounted within the cell to minimize depolarization effects that are due to window birefringence. The relative angle of the analyzer with respect to the probe beam polarizer could be precisely adjusted from outside the cell.

were mounted in rotation stages that could be precisely adjusted with micrometer screws from outside the cell by way of pressure-tight feedthroughs. Gas leakage was less than 0.08 mbar/h at 5-bars internal pressure. Despite its complexity, the present setup offers a crucial advantage for the current measurements: Because the polarizers are mounted inside the cell, any depolarization of the probe beam that is due to window birefringence is eliminated. This depolarization is caused by internal stress in the window material that varies when the internal pressure varies. A fractional change in internal pressure is sufficient to degrade the polarization quality of the beams to a level at which the sensitivity of the method is lost. Under such circumstances a proper interpretation of the pressure dependence of the signal intensity becomes impossible. Deformation of the cell caused by pressure changes could possibly lead to a misalignment between probe polarizer and analyzer and probe leakage would occur. We checked for such effects but the rigid design of the cell (stainless-steel walls, 1.2-cm wall thickness) avoided all such problems. To preserve the fixed relationship between the planes of polarization of pump and probe beams at all pressures, an additional (fixed) polarizer was mounted in the pump beam path inside the entrance cube, oriented at  $45^\circ$  to the probe polarizer. After the cell, we separated the TIPS signal from residual stray light by using a long focal length ( $f = 500\text{-nm}$ ) lens to collimate the beam before focusing it through an  $80\text{-}\mu\text{m}$  pinhole in front of the photomultiplier tube (Hamamatsu R758). The signal was collected by a gated boxcar integrator (Stanford SR250) and passed to a laboratory computer for storage and analysis.

## 3. Results

We performed an intensity check of the TIPS signal by varying, independently, both the pump and probe beam intensities  $I_p$  and  $I_{pr}$ , respectively. Two antireflection-coated quartz plates were rotated counterclockwise for this purpose, which introduced no beam walk while allowing an intensity variation over nearly two decades. The signal varied linearly with  $I_{pr}$ , as expected for a PS process. However, the signal scaled as  $I_p^{3.6}$ , revealing the high nonlinearity of the process. The findings confirm earlier mea-

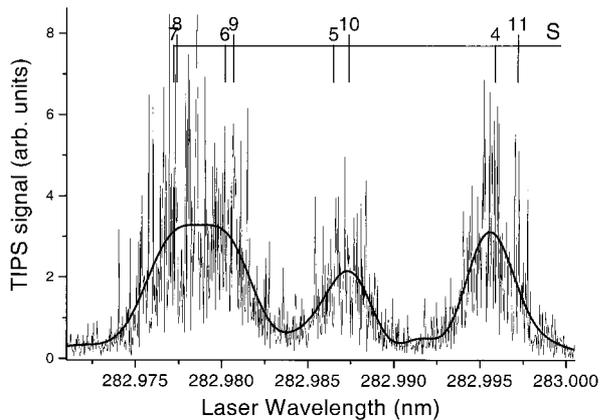


Fig. 2. TIPS S-branch bandhead in the nitrogen  $\alpha^1\Pi_g(v' = 1) \leftarrow X^1\Sigma_g(v'' = 0)$  electronic system at a total pressure of 3.01 bars. The spectrum was normalized by the averaged laser intensity profile over the scan range. The thin solid line is unprocessed raw data, indicating the enormous signal fluctuations encountered in such a high-order process. The thick solid curve is a Fourier-transformed, low-pass-filtered version of the data. Positions and rotational quantum numbers of the S-branch transitions are indicated in the upper part of the figure.

measurements<sup>24</sup> on this system in atmospheric flows of  $N_2$  where  $I_{pr}$  and  $I_p$  were varied simultaneously. Within the precision of the current measurements, this intensity behavior was independent of  $N_2$  pressure and was observed in argon-diluted nitrogen mixtures as well. For the following line shape and pressure measurements we selected a small scan range in the vicinity of the  $\alpha - X(1,0)$  bandhead of  $N_2$  with the advantage of a good signal-to-noise ratio but exhibiting partial line overlap, especially at higher pressures. Figure 2 depicts this spectral region near 283 nm of the S-branch bandhead obtained in pure  $N_2$  at 3.01 bars. The positions of the transitions probed and the corresponding rotational quantum numbers are included in Fig. 2. For these measurements the tracking system of the frequency-doubling crystal behind the dye laser was deliberately disabled because small intensity variations caused by imperfect tracking lead to enormous signal intensity fluctuations for such a high-order signal generation process. Although the beam energy did change over the scan range, the overall spectral profile was reproducible from scan to scan. The TIPS signal was normalized by the laser energy at each spectral location according to the previously determined intensity dependence. Signals were strongly attenuated with neutral density filters (optical density of 4) before being passed onto the photomultiplier tube. Above 2 bars, visible signals were obtainable when focused onto a piece of white card. Figure 2 shows the unprocessed raw data from the scan (thin line), and a smoothed spectral shape resulting from a Fourier-transformed and low-pass-filtered experimental trace (thick curve, cutoff frequency of 500 Hz). The large fluctuations of the raw signal show a disadvantage with such high-order techniques: For precise measurements, extremely stable laser sources or long

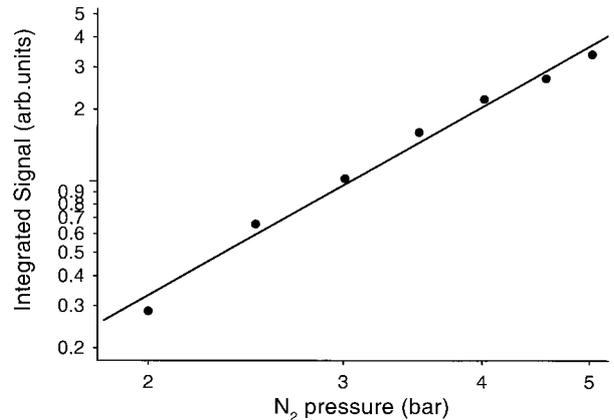


Fig. 3. Pressure dependence of the TIPS signal intensity from the pure nitrogen samples. The data points correspond to the spectrally smoothed integrated TIPS line shapes (see text) of the S(4) and S(11) transitions. The best-fit linear regression gives a power dependence of  $2.6 \pm 0.2$ .

averaging times are required. Such considerations could impose limitations on the use of the technique in dynamic measurement situations. In the present situation the problem is even more severe because of the requirement to frequency double the dye laser output, which is in itself already a second-order process.

From the smoothed spectral signatures it became evident that both intensity and linewidth of the TIPS signal increase strongly with pressure. Figure 3 presents the pressure dependence of the TIPS signal intensity for pure nitrogen on a double logarithmic plot. The filled circles correspond to the integrated area over the peaks shown in Fig. 2. For these measurements the laser energy was kept constant at 4 mJ. Error bars correspond to one standard deviation of 100 single-pulse measurements for each pressure reading. A straight line fit to the data indicates an  $N^{2.6}$  peak signal intensity behavior, where  $N$  is the number density in the probe volume. In contrast to a peak intensity measurement, the integrated line data take into account the increase with pressure of the spectral overlap between neighboring lines.

To investigate the broadening of the TIPS line with pressure, we estimated the linewidth (FWHM) of the spectral feature corresponding to the S(4) and S(11) transitions (see Fig. 2) by fitting a Gaussian line-shape function for several fixed nitrogen pressures. In the pressure range investigated in this study the results indicate a self-broadening coefficient in pure nitrogen of  $\gamma = 0.065 \pm 0.012 \text{ cm}^{-1} \text{ bar}^{-1}$ . The large error estimate takes into consideration the fact that the peak is a result of the spectral interference of two individual transitions whose lines merge at higher pressures. In addition the laser linewidth was measured and approximated by a Gaussian with a FWHM of  $0.15 \text{ cm}^{-1}$ , which is a considerable fraction of the spectral width of the transition. Two-photon line-broadening coefficients for  $N_2$  are not available, but the pressure-broadening coefficient determined

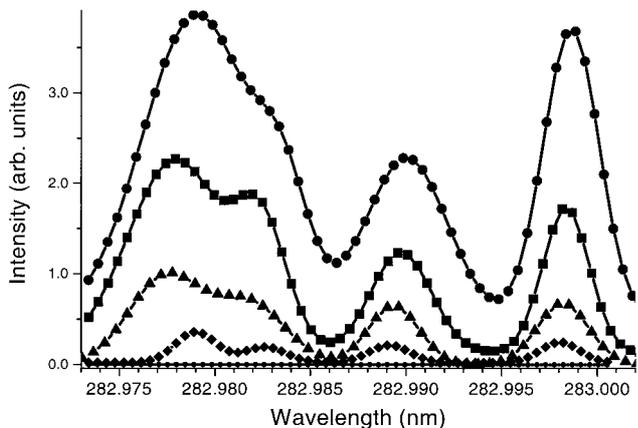


Fig. 4. TIPS *S*-branch bandhead scans (same conditions as in Fig. 2) in the nitrogen–argon mixtures at four different total pressures and a constant nitrogen partial pressure of 1.2 bars: small filled circles, 5-bars Ar only; diamonds, 1.2-bars  $N_2$ ; triangles, 1.2-bars  $N_2$  + 1-bar Ar; filled squares, 1.2-bars  $N_2$  + 2-bars Ar; large filled circles, 1.2-bars  $N_2$  + 3-bars Ar.

here is very close to that of one-photon electronic transitions of similar species, such as NO [ $2\gamma$  ( $J = 7.5$ ) =  $0.07 \text{ cm}^{-1} \text{ bar}^{-1}$ ]<sup>25</sup> in the *A*–*X* (0,0) transition. The most significant contribution in this case is thought to stem from a possible dynamic Stark effect owing to the high electric field strengths in the probe volume.

Spectral scans were also performed in mixtures that contain a constant partial pressure of  $N_2$  with argon as a variable buffer gas. Keeping the  $N_2$  pressure constant at 1.22 bars, the Ar partial pressure was changed between 1 and 4.8 bars. The result of such scans for four different pressures is shown in Fig. 4. Again, spectral smoothing was performed before the extraction of intensity and linewidth data from the experimental raw data. Surprisingly, the TIPS signal is found to increase strongly as the amount of argon in the sample increases. Furthermore, we again observed a significant broadening of the  $N_2$  line shapes with increased total pressure. The pressure-broadening coefficient of  $\gamma_c = 0.051 \pm 0.006 \text{ cm}^{-1} \text{ bar}^{-1}$  is somewhat smaller than for nitrogen perturbed by nitrogen collisions. Direct model calculations showed that an increased overlap of lines because of pressure broadening within the excitation laser bandwidth cannot account for the significant increase of the TIPS signal intensity with foreign gas pressure.

We verified that the detected signal was a true polarization signal because it disappeared completely when the pump beam was blocked or the polarizations of pump and probe beams were parallel. First it was believed that a nonresonant contribution from Ar could be the source of the increased  $N_2$  TIPS signal similar to what is observed in other nonlinear spectroscopic techniques, such as coherent anti-Stokes Raman spectroscopy.<sup>26,27</sup> However, this point was not supported by the fact that pure argon did not produce any signal at all: the signal disappeared

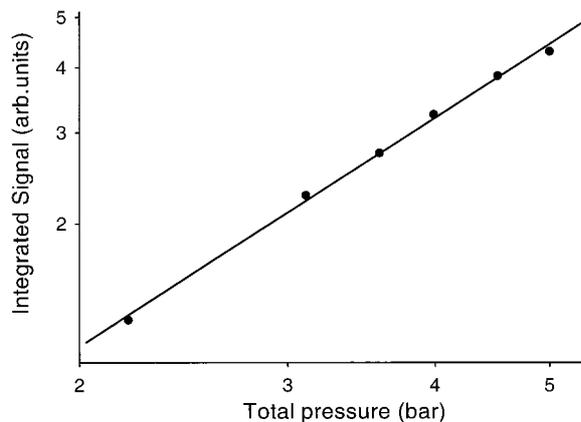


Fig. 5. Pressure dependence of the integrated TIPS signal intensity from the nitrogen–argon mixtures as a function of total pressure (the nitrogen partial pressure was 1.22 bars). Details are the same as in the Fig. 3 caption. The best-fit linear regression gives a pressure dependence of  $1.1 \pm 0.2$ .

completely when 5 bars of pure argon were used instead of the mixture (this corresponds to the trace with the small filled circles in Fig. 4). In Section 4 we provide a detailed discussion of possible mechanisms that lead to this phenomenon. Figure 5 shows the development of the integrated TIPS signal intensity in the investigated pressure range, leading to a  $p^{1.1}$  pressure dependence.

#### 4. Discussion

The dependence of the TIPS signal on the incoming laser intensities still lacks a quantitative theoretical explanation. In a simple picture of the TIPS process, where the magnetic sublevels of two resonant states are coupled by way of two-photon transitions, one would expect the signal to be proportional to the squared modulus of the third-order susceptibility  $\chi^{(3)}$  of the medium with a resulting  $I_{pr} \times I_p^2$  behavior of the TIPS signal intensity. However, it is possible that, at the high-power densities required to drive the two-photon transitions, higher-order processes (e.g.,  $\chi^{(5)}$ ) become significant, exhibiting a much stronger than  $I^3$  intensity dependence.

A second aspect that comes into play at the high pulse energies in the pump and probe beams employed in the present experiments is the question of whether a saturation of the two-photon-allowed electronic transition in nitrogen is possible in the examined pressure ranges. Assuming a  $1/e^2$  intensity contour radius of 1.5 mm at the pump beam focusing lens (250-mm focal length) the waist radius  $w_0$  of a  $TEM_{00}$  Gaussian mode at the focal point is approximately 20  $\mu\text{m}$ . In our case this gives a corresponding intensity of  $32 \text{ GW/cm}^2$  at the focal point (assuming an energy of 4 mJ in a 10-ns pulse). This value is an upper limit when we take into consideration the quality of the beam profile of the frequency-doubled dye laser beam that was far from diffraction limited.

An indication for the degree of saturation in the

two-photon PS experiment can be provided by comparing the rates for two-photon absorption into, and collisional deactivation out of, the  $a\ ^1\Pi(v' = 1)$  electronically excited state of molecular nitrogen. To the best of our knowledge, however, the absolute two-photon absorption cross section as well as collisional deactivation rates out of the excited state are not known accurately. The two-photon excitation rate ( $s^{-1}$ ) for a one-color experiment can be written as<sup>28</sup>  $W^{(2)} = \sigma^{(2)}I(\omega_1)/(\hbar\omega_1)^2$ , where  $\sigma^{(2)}$  is the two-photon absorption cross section ( $cm^4\ s$ ), and  $I(\omega_1)$  is the intensity of the pump beam at frequency  $\omega_1$ , respectively. For our estimate we used a value of  $10^{-50}\ cm^4\ s$  ( $2.4 \times 10^{-32}\ cm^4/W$ ) for  $\sigma^{(2)}$  derived by Crosley and Smith<sup>29</sup> for the  $\Sigma-\Pi$  (0,0) band of OH obtained from a direct comparison of line intensities of one- and two-photon transitions in the same band. This species is somewhat arbitrarily chosen but is available in the literature. A similar value of  $1 \times 10^{-30}\ cm^4/W$  was given for CO by Tjossem and Smyth.<sup>30</sup> Assuming this value is similar for the  $\Pi-\Sigma$  transition in  $N_2$ , we calculated  $W^{(2)} \sim 1.9 \times 10^7\ s^{-1}$  for the photon and beam energies employed in the present experiments. Concerning an estimate for the deactivation rate, van Veen *et al.*<sup>23</sup> determined a rate constant  $k$  for collisional quenching of the  $a\ ^1\Pi$  state at low pressure (13.3 mbars) of  $k = 0.7 \times 10^6\ Torr^{-1}\ s^{-1}$ , which translates to  $5.3 \times 10^8\ s^{-1}$  at atmospheric pressure. This value is of the same order of magnitude as collisional quenching of the two-photon-excited  $B$  state of CO by argon,<sup>30</sup> with a rate of  $3.4 \times 10^9\ s^{-1}$ . This simple estimate shows that, at the pressures and beam energies used in our experiments, collisional deactivation is much faster than population cycling and thus saturation of the TIPS signal is not a probable process. However, because of unknown cross sections for this excitation scheme, the above estimate includes a significant amount of uncertainty.

A clue for the pressure dependence of the integrated signal intensity can be culled from the observed intensity behavior of the TIPS signal. In any nonlinear third-order spectroscopic technique the signal intensity should depend on the square of the number density difference between the levels coupled in the fourfold photon-molecule interaction<sup>31</sup> and, for homogeneous broadening, should decrease with increased pressure at a constant partial pressure of the resonantly excited species. However, as is evident from Fig. 3, significant deviations from this square power law are observed for pure nitrogen. One explanation could be that a significant number of resonantly excited  $N_2^+$  ions are produced in our excitation scheme. The two-photon resonant absorption step to the  $a$  state of  $N_2$  near 283 nm can be followed by absorption of an additional photon at 283 nm to reach the electronic ground state of  $N_2^+$ . From this state close-lying single- and two-photon resonant transitions in  $N_2^+$  are possible, which may compete with the TIPS process that occurs in  $N_2$ . It is thus possible to generate single- and two-photon near-resonant PS processes in  $N_2^+$ , which are strongly enhanced, when-

ever a two-photon resonant transition in  $N_2$  is reached, inasmuch as ion production is a maximum in this case. This could lead to the intensity behavior observed in pure  $N_2$ . For a quantitative test of this hypothesis the relevant one- and two-photon absorption cross sections of  $N_2$  and  $N_2^+$  are required as well as the ionization cross sections for the transitions probed in  $N_2$ . At present, only a small amount of data is available to this end. However, in a previous study it was shown that the ionization rates from the relevant levels in  $N_2$  are similar to or greater than the two-photon transition strengths in the  $a\ ^1\Pi_g(v' = 1) \leftarrow X\ ^1\Sigma_g^+(v'' = 0)$  system of  $N_2$ .<sup>24</sup> We used this fact to devise a novel and sensitive method of  $N_2$  detection by resonant excitation of  $N_2$  in the Birge-Hopfield system by using two-photon absorption near 283 nm, which is followed by ionization of  $N_2$  through the absorption of three additional photons at 283 nm. The  $N_2^+$  ions are produced in the  $B\ ^2\Sigma_u^+$  state from which strong fluorescence near 400 nm in the  $B\ ^2\Sigma_u^+ - X\ ^2\Sigma_g^+$  system occurs, which can easily be detected. This technique has been termed (2 + 3) photon laser-induced fluorescence.<sup>24</sup> These points highlight the importance of ions that are generated by the high intensities required for the TIPS process.

Furthermore, Gladkov *et al.*<sup>32</sup> showed that close-lying electronic resonances in ambient ions can lead to strong increases in the nonresonant part of  $\chi^{(3)}$  of a species. On the other hand, electrons created simultaneously in the process do not contribute significantly to  $\chi^{(3)}$ . For the experiments with argon as the collider gas, the observed signal enhancement could originate from such an effect. From Fig. 5 it is obvious that the signal enhancement is related to the increase in Ar pressure, even though the added collider gas on its own does not produce any, or only a small amount of, nonresonant TIPS signal (see Fig. 4). It is possible that, whenever  $N_2$  is resonantly excited, such an enhancement in the nonresonant susceptibility of Ar atoms or ions (produced by electron collisions) is effected by the production of a large number of  $N_2^+$  ions that could account for the observed signal enhancement with argon pressure. Although error bars (signal fluctuations) are large, our results (compare Figs. 3 and 5) indicate that the increase of the signal with pressure is less pronounced if argon is used instead of pure nitrogen.

Finally, it was brought to our attention that the present experimental results could also be explained by the pressure-induced signal enhancements observed in multibeam multifrequency schemes such as pressure-induced extra resonances in four-wave mixing<sup>33</sup> and coherent control<sup>34</sup> in molecular systems.

## 5. Conclusions

There are obvious advantages of multiphoton polarization spectroscopy for diagnostic applications at and above atmospheric pressure. These include the almost quench-free nature of the signal intensity and a relatively straightforward experimental realization for a nonintrusive and spatially resolved measure-

ment technique. Another advantage is access to transitions that would otherwise require radiation in the vacuum ultraviolet. We have reported on the investigation of two-photon-induced polarization spectroscopy (TIPS) in  $N_2$  at elevated pressures. Studies were carried out in a high-pressure cell with an internally mounted polarizer and analyzer to eliminate the effects associated with window birefringence. The results indicate that photoionization effects play an important role in the TIPS process, which may lead to competing effects and cause an apparent enhancement of the signal. Unexpected behavior was found when Ar was added to a constant partial pressure of  $N_2$ , which led to a rise in the  $N_2$  TIPS signal as a function of Ar buffer pressure. It is possible that the presence of  $N_2^+$  ions leads to an enhancement of the single-photon nonresonant susceptibility of Ar or  $Ar^+$ . The advantage of the relative experimental simplicity of TIPS experiments is currently offset by the lack of theoretical investigations of the underlying complex physical phenomena. It is hoped that the results presented here will stimulate further theoretical interest in these phenomena. For a more accurate calculation of the populations transferred during the excitation process into the excited state with subsequent generation of ions, the solution of a rate equation model similar to that outlined by Mallard *et al.*<sup>35</sup> needs to be employed. This was beyond the scope of our experimental study but could be useful for a future quantitative understanding of the TIPS process in nitrogen.

With our data, it was not possible for us to estimate a detection limit for nitrogen by using TIPS with the present excitation scheme. However, with the double-logarithmic plot in Fig. 5 one can conclude that, with the present laser intensities, a nitrogen partial pressure of  $10^4$  Pa might readily be detectable in a mixture with argon.

Future investigations should include the effect of pressure and gas composition on  $N_2^+(2 + 3)$  laser-induced fluorescence signals that are detected simultaneously with TIPS, as well as TIPS from other species. An example is  $CO_2$  for which we observed strong, nonresonant TIPS signals in the 283-nm region.

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