Third-harmonic generation at atmospheric pressure in methane by use of intense femtosecond pulses in the tight-focusing limit

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An experimental study of third-harmonic generation in methane with a 100-fs, 820-nm, Ti:sapphire laser in a tight focusing geometry is presented. The harmonic intensity and bandwidth were measured in a range of intensities extending from below to far above the first ionization threshold and at pressures as high as 10 atm (1 atm=760 Torr). The harmonic signal follows a power-law dependence on the laser intensity with an exponent of ~7 and saturates at an intensity $I_s \sim 4 \times 10^{14}\,\mathrm{W/cm^2}$. The conversion efficiency was found to increase with the pressure for laser intensities smaller than the saturation intensity I_s and to decrease with the pressure at larger intensities. At laser intensities larger than the saturation intensity a substantial modification in the third-harmonic bandwidth and structure was observed. © 1999 Optical Society of America [S0740-3224(99)00905-4]

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1. INTRODUCTION

The generation of high-order harmonic radiation during the interaction of an intense laser pulse with a gas has been intensively studied both experimentally[1–8] and theoretically[9–17] during the past decade and is now an important source of coherent extreme UV radiation. Very high harmonic orders have been observed,[5–8] demonstrating the possibility of generating coherent light pulses with photon energies as high as 150 eV. It is well established now that, after the expected rapid decrease for the first orders, the harmonic spectrum forms a plateau of odd-integer harmonics of the incident laser frequency, extending to a maximum photon energy of approximately $I_p + 3U_p$. I_p is the free-field ionization potential of the atomic species and $U_p = (e^2E^2/4m\omega^2)$ is the ponderomotive energy (e and m are the electron charge and mass; E and ω are the laser electric field and frequency). Recent experimental studies have measured the several characteristics of the harmonic beams, such as angular distributions,[18] temporal profile,[19] and bandwidth.[6,20–23]

The theory of high-harmonic generation has been extensively developed and investigated, including the single-atom response, propagation, and phase-matching phenomena. Several methods have been used to describe the atomic response. One of the most comprehensive treatments of high-field harmonic generation is that reported by L'Huillier *et al.*[24] In that study the response of a single atom to the laser field is determined by the integration of the time-dependent Schrödinger equation by use of a frozen core potential approximation. This model includes a single-valence electron in the field of an effective core potential, the remaining valence electrons frozen in their ground-state orbital, and a linearly polarized field. The model was used to calculate the dynamic polarizabilities and harmonic spectra in strong fields. Another prominent method used to describe the atomic response is solution of the coupled time-dependent Floquet equations for hydrogen.[25] A number of simplified models of high-harmonic generation were also used, such as one-dimensional approximations in a soft-Coulomb potential,[26] the integration of the equations of motion for a classical hydrogen system,[27] two-level system models in a strong field,[14,28–30] and a slowly driven anharmonic oscillator.[16] The majority of the studies of harmonic generation during the interaction of strong fields with gases dealt with harmonics in the plateau region. Measurements of low-order harmonics were performed mainly before the advent of terawatt laser sources. Studies of frequency tripling revealed a harmonic behavior well described by the lowest-order perturbation theory.[31] Several studies of harmonic generation in the strong-field regime investigated low-order harmonics. Fifth-harmonic production in neon and argon irradiated with a 248-nm picosecond laser at focused intensities in the range from 10^{13} to $10^{15}\,\mathrm{W/cm^2}$ was reported in Ref. 32. The pressure and intensity dependence of the seventh harmonic in xenon at intensities as much as $2 \times 10^{14}\,\mathrm{W/cm^2}$ was studied in Ref. 33. The third harmonic in xenon produced with a 1064-nm Nd:YAG laser at irradiances as high as $5 \times 10^{13}\,\mathrm{W/cm^2}$ was studied in Ref. 34.

There are few reports of harmonic generation in the

strong-field regime in a tight-focusing[32–34] geometry. The majority of the harmonic-generation experiments were performed in a loosely focused geometry, required for phase matching in a positively dispersive medium and useful for avoiding absorption of the harmonic radiation. Most of the harmonic generation studies in gases were performed with rare gases used as the nonlinear medium.[1–6] Other experiments used molecular gases, such as hydrogen and nitrogen[33,35] and solid alkali metal targets.[36] A survey study of high-order harmonic generation in molecular gases was also reported.[37]

Recently there has been an increasing interest in third-harmonic generation in gases with ultrashort pulses as a source of UV pulses. Calculations by Vanin *et al.*[38] showed that very short UV pulses (<20 fs) can be generated with an efficiency of 1%. Alternative schemes for generating short UV pulses, such as frequency mixing in crystals, result in much longer pulse durations because of dispersion and bandwidth limitations. Measurements of spectral and propagation characteristics of third-harmonic pulses generated by 1.5-mJ, 80-fs, 800-nm laser pulses in subatmospheric pressure noble gases were reported by Backus *et al.*[21] Blueshifted third-harmonic generation and correlated self-guiding during ultrafast barrier-suppression ionization of subatmospheric density noble gases were performed by Siders *et al.*[22] Spectral spatial measurements of fundamental and third-harmonic light of intense 25-fs laser pulses focused in a gas cell were reported by Peatross *et al.*[23]

In this paper we report measurements of the third-harmonic generation in methane in the range of laser irradiances extending from below to far above the ionization threshold. We studied the variation of third-harmonic intensity and its bandwidth as a function of the laser irradiance and the gas pressure. The experiments reported here were performed in a tight-focusing geometry, in which the laser confocal parameter b is smaller than the length of the nonlinear medium. We extended our studies to pressures as high as 10 atm (1 atm: 760 Torr) and also examined the pressure dependence of the bandwidth of the third harmonic.

2. EXPERIMENT

The third-harmonic generation experiments consisted of focusing an intense laser pulse into a nonlinear gas medium and then measuring, along the propagation axis, the UV light emitted during the interaction. The laser used in the experiments was a Ti:sapphire laser system based on chirped pulse amplification. The oscillator is a Tsunami pumped by a 5-W, 514-nm argon laser. This oscillator produces 70-fs pulses with an energy of 5 nJ/pulse at a wavelength tuneable from 700 to 850 nm. The pulse is stretched to 1.5 ns and amplified in a regenerative amplifier followed by a four-pass Ti:sapphire amplifier, pumped by a Nd:YAG laser, to 75 mJ. After compression the laser delivers 100-fs pulses at a 10-Hz repetition rate and energy as high as 50 mJ/pulse. In these experiments we operated the laser at a central wavelength of 826 nm.

The laser beam was focused with an achromatic 80-mm focal-length lens into a 20-cm-long gas-filled cell. The confocal parameter $b = 2\pi\omega_o^2/\lambda$ (with ω_o denoting the beam radius and λ the wavelength) was roughly 60 times smaller than the length of the cell. A schematic description of the experimental setup is shown in Fig. 1. The laser intensity was measured with a powermeter mounted at the input end of the cell. A thin glass window was situated at the input end of the cell, and a quartz window at the output end, transparent to UV radiation. This output radiation passes through two mirrors with a reflectivity of 99.9% for 820 nm, mounted after the output end of the cell and reflecting the fundamental light. A monochromator, McPhearson Instrument Model 2051 with a 1200-groove/cm plane grating, placed after the two mirrors is used to select the third harmonic. The spectral resolution of the monochromator was 0.05 nm. Three detectors were used to measure the signal: a CCD camera, an optical multichannel analyzer, and a photomultiplier. A computer-based data-acquisition system was used to record the signal. Gas targets of CH_4 were provided at different densities by changing the pressure in the gas cell between 1 and 10 atm. To ensure that the collected light was not laser-induced fluorescence, we translated the monochromator perpendicular to the beam propagation direction. All the UV light was found to be strongly directional, propagating collinearly with the fundamental beam. The experimental setup was designed so that the laser intensity was not high enough to induce nonlinear optical effects in the cell's windows.

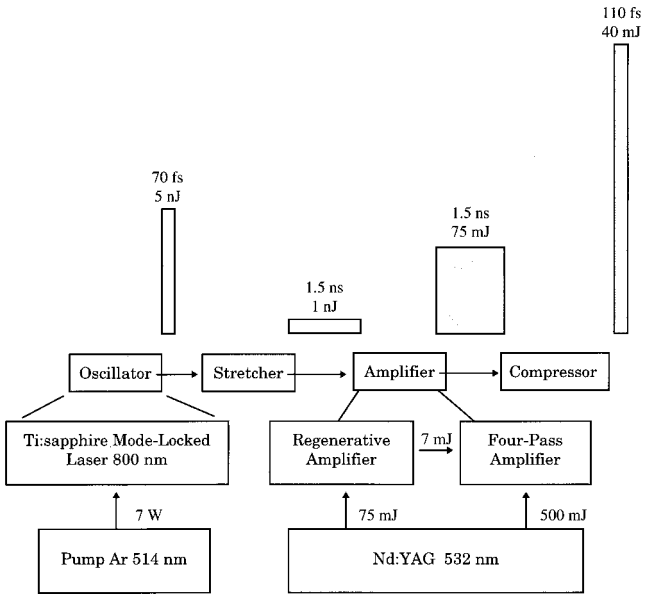

Fig. 1. Schematic of the experimental setup for measuring the third harmonic.

3. EXPERIMENTAL RESULTS

In the experiments we measured the third-harmonic intensity and bandwidth as functions of the laser irradiance and the gas pressure. The third-harmonic intensity as a function of the laser intensity in the range 4×10^{13} to 2×10^{15} W/cm^2, at a gas pressure of 30 psi (1 psi $\approx$ 53.2 Torr), is shown in Fig. 2. Each data point plotted represents the mean value of 32 shots, and the variation from shot to shot was less than 20%. Most of the data were taken at laser intensities above 8×10^{13}

W/cm^2, higher than the intensity corresponding to the first ionization threshold[32] in methane. The third-harmonic signal follows a near power-law dependence with an exponent of 7.1. Above $I_s \sim 4 \times 10^{14}$ W/cm^2, we observed a much slower exponential growth, of ~1.6. The saturation of the signal at the same laser intensity shown in Fig. 2 was also seen at the higher pressures studied here.

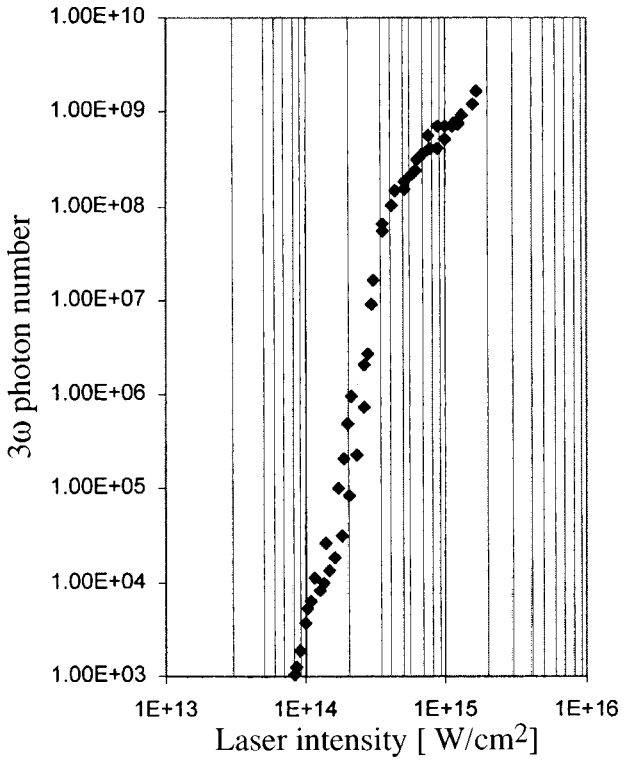

Fig. 2. Third-harmonic signal in CH_4 as a function of the laser intensity at various gas pressures.

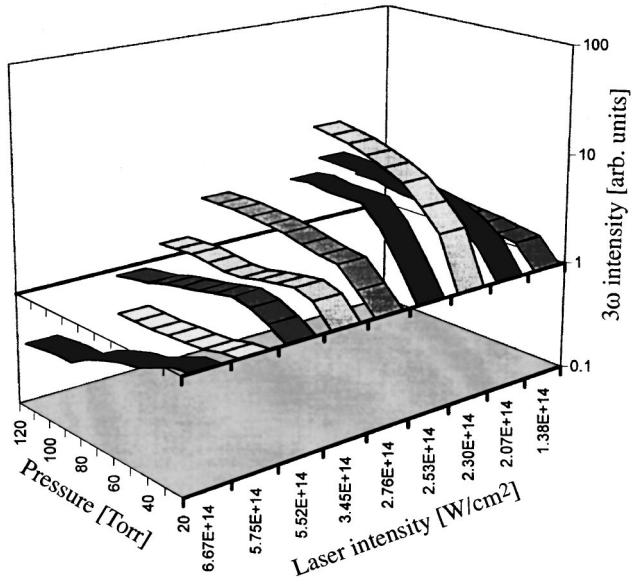

Fig. 3. Intensity of the third harmonic radiation in CH_4 at different values of the laser irradiance as a function of pressure.

The third-harmonic signal as a function of pressure, for several different intensities in the range 1.4×10^{14} to 10^{15} W/cm^2, is shown in Fig. 3. The measured pressure dependence is weaker than the square pressure dependence reported in other studies.[10,13] As the intensity increases, a plateau appears at high pressures. At laser intensities of the order of 3.5×10^{14} W/cm^2, the harmonic signal rises with increasing pressure, reaches a maximum at a pressure of ~30–40 psi, and then decreases. For laser intensities higher than 4×10^{14} W/cm^2, the harmonic signal decreases as the pressure increases.

The shape and the bandwidth of the third-harmonic signal were measured as a function of the laser intensity and the gas pressure. Figure 4 depicts the spectra of the radiation emitted in the vicinity of 275.3 nm at a gas pressure of 30 psi for several different laser intensities. At the low laser intensity the spectrum is asymmetric, displaying a peak near 274.5–274.7 nm with a width of ~1.2 nm. As the laser irradiance increases, the radiation spectrum changes in a complicated manner. The asymmetry is higher, with several peaks appearing at shorter wavelengths. At $I = 1.7 \times 10^{14}$ W/cm^2, there is a peak at 274.9 nm, with a width of 10–12 nm. A shoulder feature is also seen, extending from 273.2 nm to 274.0 nm. At $I = 2.46 \times 10^{14}$ W/cm^2, there is a peak at 275.2 nm, with a width of ~0.7 nm, a pedestal extending from 273.7 to 274.5 nm and a peak at 273.1 nm with a width of 4 nm. At higher laser intensities, three or four peaks are distinguished at wavelengths from 272.5 to 275.8 nm. The total bandwidth of the harmonic signal increases with the laser intensity and reaches ~3.0 nm. The jagged peaks seen in the spectrum of the third harmonic were present in the fundamental beam.

Figure 5 displays the spectra of the harmonic signal at various gas pressures in the range 20–100 psi, measured at a laser intensity of 1.1×10^{14} W/cm^2, lower than the saturation intensity I_s. At the lowest pressure displayed in Fig. 5, the center of the harmonic signal is at 274.6 nm. The spectrum develops a pedestal at 276.0 nm. At higher intensities this pedestal develops into a distinct peak. The blueshift of this peak is a nonlinear function of pressure. Figure 6 shows the spectra of the third-harmonic signal at various gas pressures, measured at a laser intensity higher than I_s. Again it is seen that the spectrum depends on the gas pressure in a rather complicated way. The harmonic signal is split into three to five peaks. Additional peaks are measured at 275.5, 274.2, 273.8, and 273.3 nm. A pedestal extending from 275.5 to 276.0 nm is seen at high pressures. The overall signal is broadened. Therefore it is found that at laser irradiances larger than the saturation irradiance the third-harmonic structure and bandwidth become pressure dependent.

For comparison we also measured the third-harmonic signal in two noble gases in the same range of laser intensity and pressure. The intensity dependence of the third harmonic in argon, xenon, and methane measured at 30 psi is displayed in Fig. 7. The third-harmonic signal in xenon follows a power law with an exponent of 4 followed by a rapid change in slope at 2×10^{14} W/cm^2, whereas the harmonic signal in argon is proportional to $I_1{}^9$ up to intensities of 4×10^{14} W/cm^2, and then saturates.

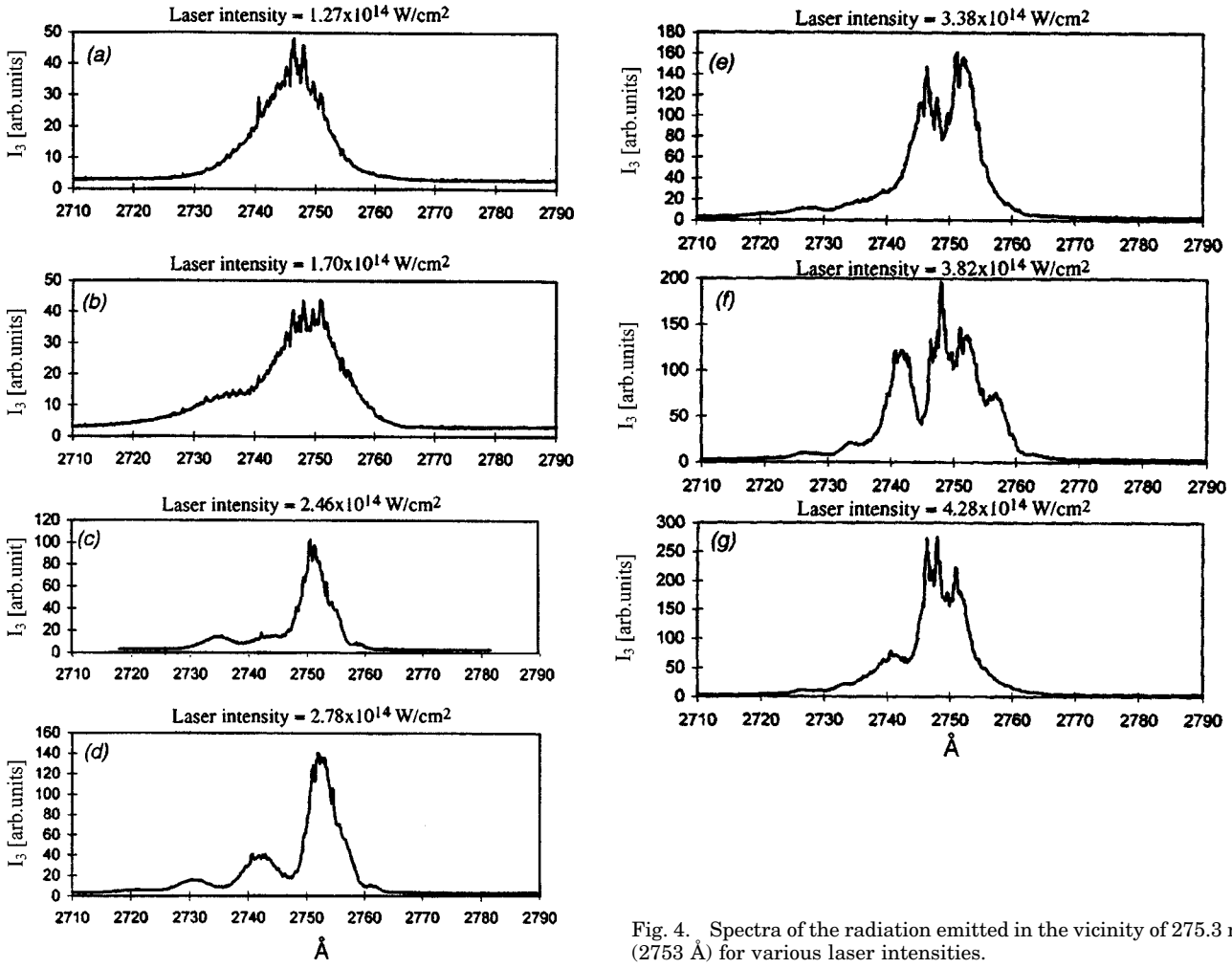

Fig. 4. Spectra of the radiation emitted in the vicinity of 275.3 nm (2753 Å) for various laser intensities.

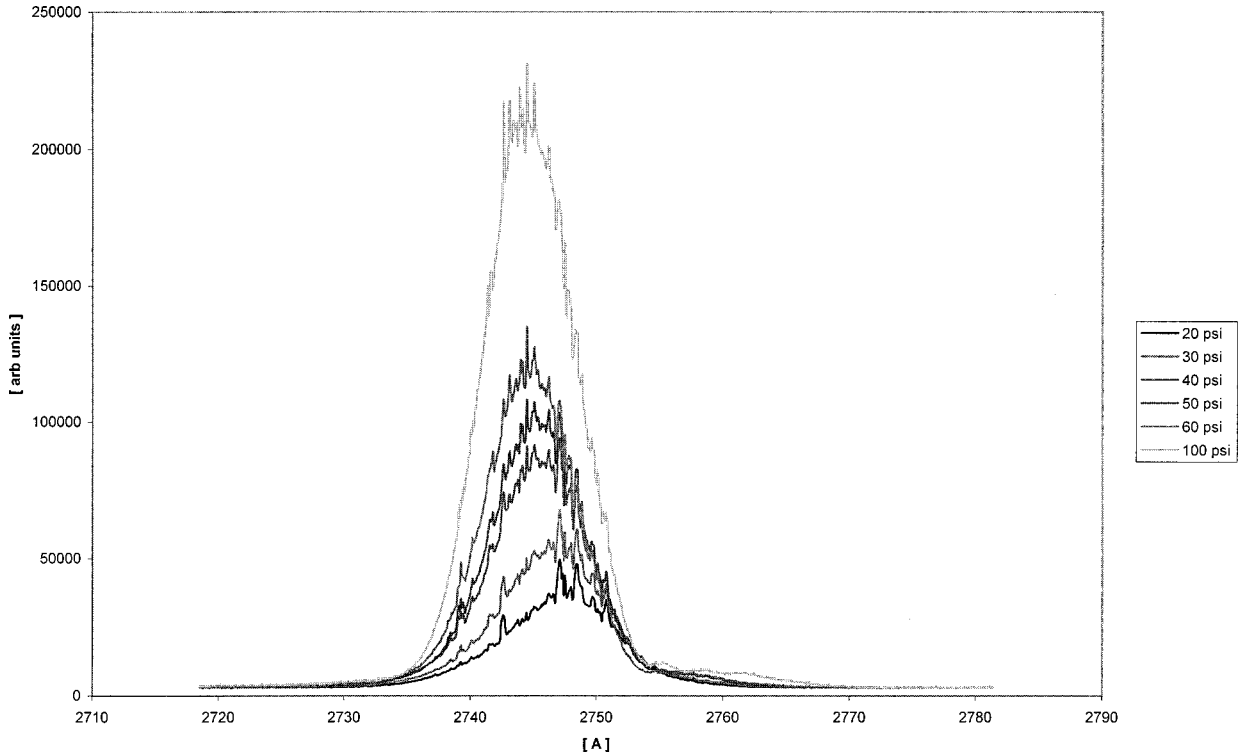

Fig. 5. Spectra of the third-harmonic at various gas pressures, measured at a laser intensity $I = 1.15 \times 10^{14}$ W/cm^2.

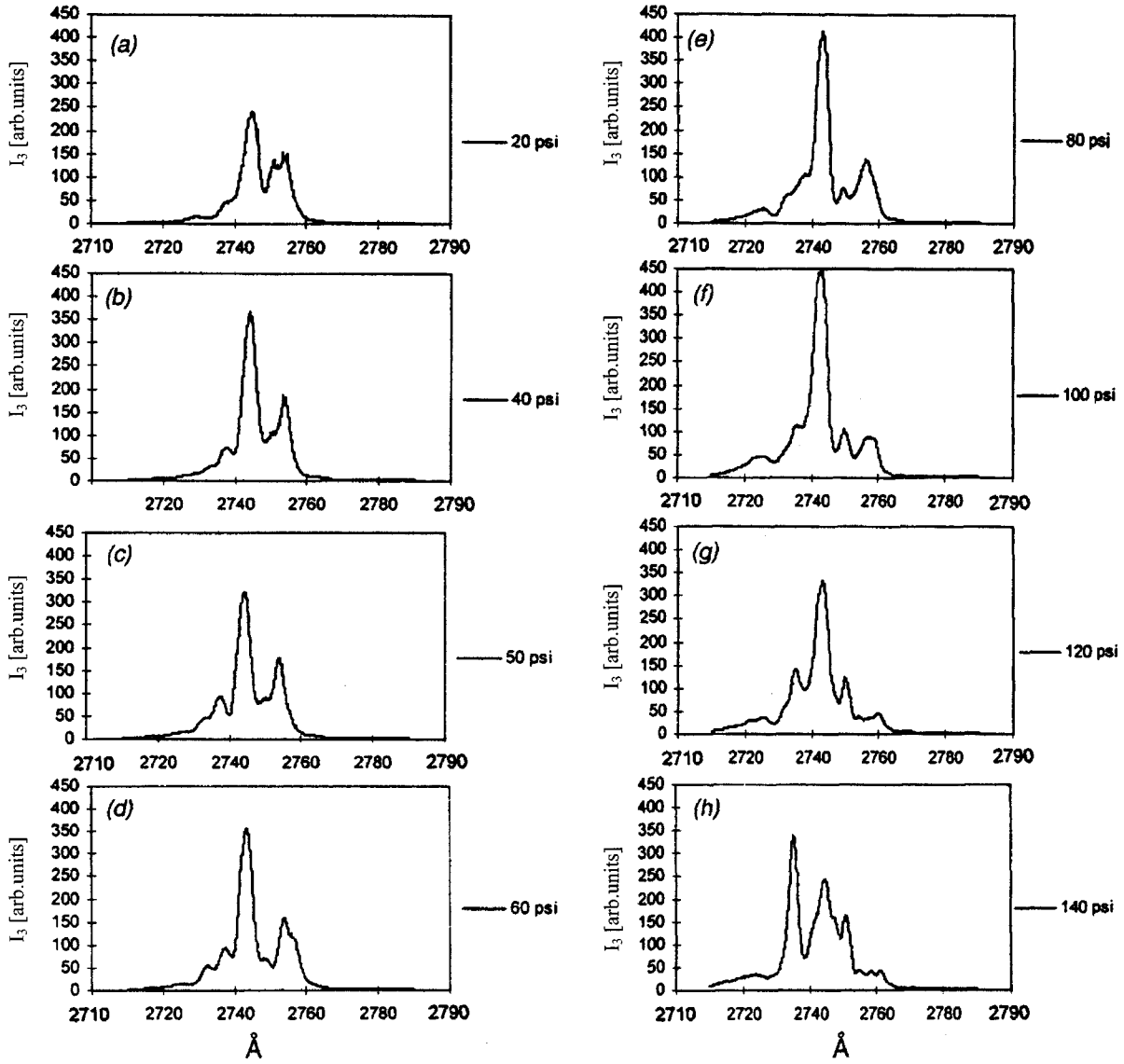

Fig. 6. Spectra of the third harmonic at various gas pressures, measured at a laser intensity $I = 4.5 \times 10^{14}\,\mathrm{W/cm^2}$.

4. INTERPRETATION OF EXPERIMENTAL RESULTS

A complete theoretical treatment of the experimental results would be a very complicated task. The full theory of harmonic generation must include the complete atomic level structure of the target atoms, the nonperturbative interaction of these levels with the intense laser field, the depletion of the neutral atoms due to ionization, the dispersion of the resulting plasma, the geometrical complications resulting from the spatial and the temporal distribution of the field strengths near the focus, and the propagation of the fundamental and the harmonic waves through the perturbed medium. There is no currently available theoretical technique that can handle all of these effects simultaneously. Low-order harmonic generation has usually been described by perturbation theory. An excellent description of this approach is given in Ref. 39. The recent paper by Vanin *et al.*[38] studied low-order harmonic generation in gases, at intensities in the range 10^{15}–$10^{16}\,\mathrm{W/cm^2}$, corresponding to tunneling and barrier-suppression ionization. Vanin *et al.* solve a one-dimensional wave equation for a laser pulse coupled by a nonlinear polarization to a one-dimensional Schrödinger equation for a one-electron atom, taking into account ionization, harmonic generation, and adiabatic blueshifting of the laser radiation. The model predicts ultrafast ($\leqslant$20 fs) pulses of harmonic generation, frequency broadened and blueshifted. Our experimental results were done in a lower laser intensity range, 5×10^{13} to $2 \times 10^{15}\,\mathrm{W/cm^2}$, when tunneling and multiphoton ionization take place. The opinion prevailing for this laser intensity regime is that significant ionization hinders efficient generation of the harmonics. Because there is no currently available theoretical technique that takes into account all the effects of the laser–gas interaction, we first compare the experimental results with a simple perturbative description that neglects many of the topics mentioned above.

Assuming that the incident electric field has a Gaussian spatial profile, with a confocal parameter b

$= 2\pi\omega_o{}^2/\lambda$, the harmonic wave is also a Gaussian with the same confocal parameter. The intensity of the third harmonic I_3 is given by

$$I_3 \sim {I_1}^3|\chi^{(3)}|^2N^2|F_3(b\Delta k)|^2, \tag{1}$$

where $\chi^{(3)}$ is the third-order susceptibility per atom, N is the gas number density, and $F_3(\Delta k)$ is the phase optimization integral, which may be approximated[31,39] in the tight-focusing limit as

$$F_3(b\Delta k) = 2\pi\frac{b\Delta k}{2}\exp\left(\frac{b\Delta k}{2}\right), \qquad \Delta k < 0,$$

$$F_3(b\Delta k) = 0, \qquad \Delta k > 0. \tag{2}$$

Here Δk is the wave-vector mismatch, which is related to the index of refraction according to

$$\Delta k = 3\frac{2\pi}{\lambda}(n_3 - n_1), \qquad n_3 - n_1 = 2\pi N\,\mathrm{Re}[\chi^{(1)}(3\omega)]. \tag{3}$$

According to Eqs. (2), harmonic generation described by the lowest-order perturbative model is possible in the tight-focusing limit only in media with negative Δk. From Eq. (1) it is seen that the third-harmonic signal should be proportional to the third power of the fundamental laser intensity. The signal should increase with the density, reach a maximum, and then decrease at high densities.

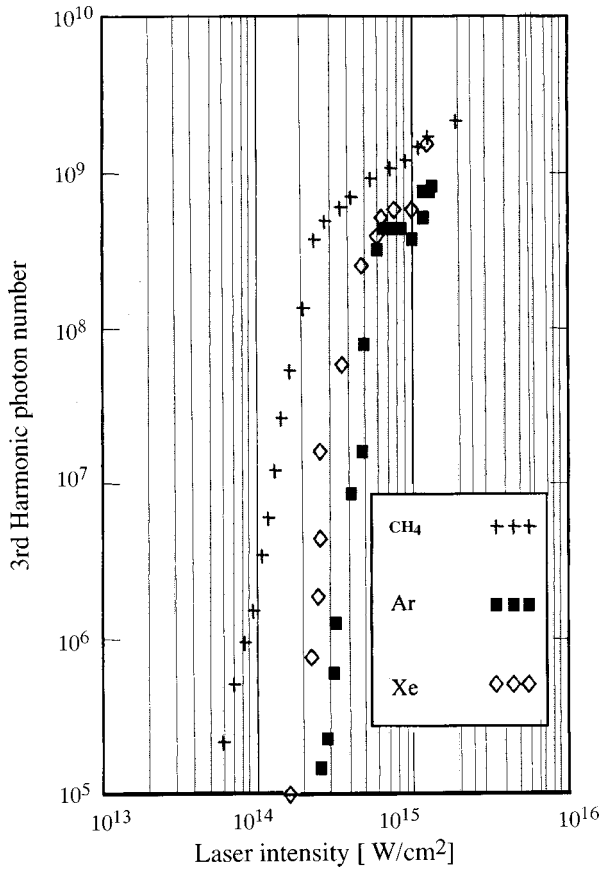

Fig. 7. Harmonic signal in xenon, argon, and methane measured at 30 psi as a function of the laser intensity.

Our experimental data exhibit power-law intensity dependence with a higher exponent, 7.1, than is predicted by Eq. (1).

A calculation of the index of refraction of methane would involve the knowledge of all its energy levels together with their shifts induced by the interaction with the laser and is beyond the scope of this study. Calculations[40] show that the index of refraction of argon and xenon near 253 nm is positive. Third-harmonic generation can occur, however, in a gas cell if the laser propagation is modified during the interaction with the gas, if the third-harmonic generation deviates from the perturbative law,[34,41] or if Δk has nonlinear components.

In what follows we consider several processes that may affect the harmonic production and shed light on the deviation of the experimental data from the perturbative model. A mechanism that may explain the signal intensity dependence and the decrease with pressure at high intensities, found in the experiments, is the nonlinear index of refraction.[32,33,41] The nonlinear index of refraction can affect the frequency conversion process by introducing intensity-dependent phase shifts, between the fundamental and the harmonic waves, that alter phase matching. An intensity-dependent Δk may change the intensity and pressure dependence given in Eqs. (1) and (2).[32,33,41]

An effective positive nonlinear index of refraction may lead to self-focusing of the fundamental beam, change its beam profile and propagation, and affect the third-harmonic generation. The critical power for self-focusing is

$$W_{\mathrm{cr}} = \frac{(1.22\lambda)^2c}{256N\chi^3}, \tag{4}$$

where χ^3 is the third-order susceptibility per atom. Using the value $\chi^3 = 0.8 \times 10^{-38}\,\mathrm{cm}^6/\mathrm{erg}$,[42] the critical power for self-focusing at 30 psi is $W_{\mathrm{cr}} \approx 2 \times 10^{11}\,\mathrm{W}$, of the order of the laser power in the experiments here.

Another process that may affect third-harmonic generation is the ionization of the medium during interaction with the laser field. The neutral gas is depleted in a nonuniform matter, changing the density of the medium. The ions created may themselves contribute to harmonic generation, probably less efficiently than the neutrals. The free electrons change the refractive index, and their contribution to harmonic generation is twofold. First, they change the propagation of the incident beam, introducing a defocusing. Second, the electrons contribute to the mismatch vector, affecting the phase-matching conditions. The change in the refractive index $\Delta n_q{}^e = -\omega_p{}^2/2q^2\omega^2$ is space and time dependent, following the variation of the electron density; q is the harmonic order and $\omega_p = (4\pi N_e e^2/m)^{1/2}$ is the plasma frequency. Defocusing becomes significant when the defocusing length $l_D = (\lambda/2)(N_{\mathrm{cr}}/N_e)$ is shorter than the Rayleigh length[43] $z_R = (\pi\omega_o{}^2/\lambda)$. N_e is the electron density, and $N_{\mathrm{cr}} = (1.1 \times 10^{21})\lambda\,\mu\mathrm{m}^{-2}\,\mathrm{cm}^{-3}$ is the critical density. Therefore the condition for defocusing is

$$\frac{N_e}{N_{\mathrm{cr}}} \geqslant \frac{\lambda}{2z_R}. \tag{5}$$

Using inequality (5) for our experimental conditions, the electron density for defocusing should be $N_e \sim 1.3 \times 10^{18}\,\mathrm{cm}^{-3}$. This value corresponds to entire medium ionization at a pressure of 1 psi, which is lower than the pressures studied here.

Moreover, owing to the decrease in the refractive index induced by plasma formation during the laser pulse, a blueshift of the fundamental and of the harmonics is expected. The spectral shift of the third harmonic originates from the spectral shift of the fundamental and from the variation of the refractive index at frequency 3ω. The frequency shift of the fundamental is given by

$$\frac{\Delta\lambda}{\lambda} = \frac{z_R}{c}\frac{\mathrm{d}n}{\mathrm{d}t}, \tag{6}$$

where $n = [1 - {\omega_p}^2(t)/\omega^2]^{1/2}$ is the plasma index of refraction. For the third-harmonic wavelength and assuming that the electron density changes from 0 to $10^{17}\,\mathrm{cm}^{-3}$ during 100 fs, the expected blueshift is ~1.0 nm, in approximate agreement with the observed frequency shift. A blueshift of the third-harmonic signal generated during ultrafast ionization in noble gases and air was predicted by Vanin *et al.* and reported recently.[21–23]

Focusing and defocusing of the laser beam may occur at such intensities and affect the generation of the harmonics. It was shown recently that the interplay of self-focusing due to a nonlinear index of refraction and defocusing due to ionization affects the propagation of an intense laser beam in gaseous medium. Nibbering *et al.*[44] studied the beam distortions of a 150-fs, 800-ns pulse as it is propagated 40 m through air. They observed self-focusing to the point that an initially collimated beam converged to reach intensities near $10^{14}\,\mathrm{W/cm}^2$, forming a channel over many meters owing to a quasi balance between Kerr focusing and the defocusing arising from a small degree of ionization. A balance of, or unstable oscillation between, self-focusing and defocusing during third-harmonic generation in noble gases was reported also by Siders *et al.*[22]

To test whether self-focusing due to a nonlinear index of refraction or defocusing due to ionization occurred in our experiments, we measured the beam diameter in the gas as a function of the laser intensity. The beam diameter did not change when the laser intensity was increased. Since for the laser intensities and pressures used in our experiments both defocusing and self-focusing should take place, a quasi balance of these two processes might have occurred. In addition to frequency shift and broadening, at laser intensities higher than the saturation intensity the harmonic signal exhibits a general trend of increasing complexity as a function of pressure. This feature is characteristic of self-phase modulation.[45] The interplay of self-focusing, defocusing, and self-phase modulation, which induces spectral broadening and splitting of the fundamental, could lead to a decrease in the peak power and therefore to a decrease in third-harmonic generation. However, in our experiments we did not observe a significant modification in the structure of the spectrum of the fundamental beam.

Another explanation for the observed third-harmonic signal intensity scaling law is a high-order process. If the simple harmonic generation ($\omega + \omega + \omega \rightarrow 3\omega$) is forbidden, the next-higher-order process could begin to play a role. The next-order process is six-wave mixing ($\omega + \omega + \omega + \omega \rightarrow \omega + 3\omega$), i.e., absorption of four ω photons and emission of one ω photon and one 3ω photon. However this process does not explain the ${I_1}^7$ dependence. An eight-wave process, ($\omega + \omega + \omega + \omega + \omega \rightarrow \omega + \omega + 3\omega$) may explain the intensity dependence.

Another effect that may influence the harmonic generation conversion efficiency is a modification of the single-atom response at high densities.[8] Harmonic generation in the low-frequency, high-intensity regime can be described as a semiclassical two-step model[10,46]: The electron escapes the atomic core by tunneling through the suppressed barrier of the Coulomb potential, which is modified by the laser electric field. Free of the atomic potential, the electron oscillates classically in the laser field and then comes back with a high energy toward the nucleus; the nucleus and the electron may recombine to the ground state of the atom, giving up the kinetic energy as a harmonic photon. The quiver amplitude of an electron exposed to an intensity of $10^{15}\,\mathrm{W/cm}^2$ at 0.820 μm is 1.8 nm. At the highest pressures used in our experiments, ~10 atm, the average distance between two molecules is 2.3 nm, almost the same as the quiver amplitude. Therefore it may be expected that the return of the electron toward the nucleus is affected by the presence of the other molecules.

5. CONCLUSION

We measured third-harmonic production in methane by using a 100-fs, 826-nm, Ti:sapphire laser. The experiments were performed in a tight-focusing geometry, at laser intensities ranging from below to far above the first ionization threshold in methane, and pressures ranging from 20 to 140 psi. It was found that the third-harmonic signal response is different above and below $I_s = 4 \times 10^{14}\,\mathrm{W/cm}^2$. (1) The third-harmonic signal follows an ${I_1}^7$ law and then saturates at an intensity I_s (Fig. 2). (2) At laser intensities smaller than the saturation intensity I_s, the harmonic signal increases when the pressure is increased, whereas at intensities above I_s the signal decreases with increased pressure (Fig. 3). (3) The dependence of the third-harmonic spectrum on the pressure is also different when the laser intensity is lower or higher than the saturation intensity. At intensities below I_s the structure of the third-harmonic spectrum did not change significantly when the pressure was changed (Fig. 5). However, at laser intensities above I_s, the structure of the spectrum was highly sensitive to pressure variation (Fig. 6).

Several mechanisms were proposed for the interpretation of the experimental data: (1) a nonlinear index of refraction, (2) a process higher than third order, or (3) changes in the propagation of the fundamental beam caused by self-focusing or defocusing. Focusing and defocusing effects may be responsible for the I^7 power law as measured and for the saturation. However, they fail to explain the two other phenomena. Moreover, we did not observe any significant pressure tuning of the fundamental beam spectrum that might indicate an interplay of self-focusing and defocusing. A high-order process

may explain the I_1^7 scaling law and the saturation of the harmonic signal and the modifications in the spectra structure but does not explain the pressure dependence. The nonlinear Δk changes in the wave-vector mismatch due to laser intensity and pressure variations may be the reason for both the scaling law and saturation and the pressure dependence. Therefore nonlinear changes in Δk provides the favored explanation for the above results. The increased complexity of the harmonic spectrum as a function of pressure was seen in other studies.[42,47,48]

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