Spectral-spatial measurements of fundamental and third-harmonic light of intense 25-fs laser pulses focused in a gas cell

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We present spatially and spectrally resolved measurements of light emerging from the focus of an intense 25-fs laser pulse in gaseous media. The pulses are focused inside a chamber backfilled with either argon or air to nominal vacuum intensities of up to 5×10^{15} W/cm^2, far in excess of that necessary for ionization. We have recorded spectral-spatial maps for various pressures and pulse energies and have compared the results of argon and air. This survey is done in the regime where ~0.1% of the laser light is converted into the third harmonic, which is of practical importance since it is the largest conversion efficiency measured to date while maintaining such a short pulse duration. These measurements are, to our knowledge, the first of their type and are performed with a laser beam of very high spatial quality. The dynamics of an intense laser focusing into a gas cell is complicated, and these measurements provide a way to compare numerical simulations with an array of experimental information. The experiments also determine the conditions for optimal third-harmonic spatial beam quality in this regime. © 1997 Optical Society of America [S0740-3224(97)02112-7]

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1. INTRODUCTION

We recently reported[1] a ~0.1% efficiency for converting our 800-nm, 25-fs laser pulses[2] into the third harmonic at 267 nm simply by focusing 1-mJ pulses in air or argon (backfilled chamber). Most importantly, the UV pulses were found to be very short in duration (sub-20 fs). Alternative schemes of converting laser light into UV light, such as frequency mixing in crystals, result in much longer pulse durations because of dispersion and bandwidth limitations. For our laser conditions, we found third-harmonic conversion to be most efficient when focusing in a relatively loose geometry (*f*/30–*f*/40). During the interaction, a plasma is produced in the focus, and the laser beam undergoes blueshifting and spatial distortions as ionization of the gas takes place. The goal of the present work is to gain insight into this complex interaction (i.e., propagation and distortion of the fundamental as well as the generation and propagation of third-harmonic light) through a study of the spectral and spatial properties of the emerging light.

In the low-intensity or perturbative limit, third-harmonic conversion of laser light cannot take place for a Gaussian laser beam focused in a cell of positively dispersive gas.[3–7] This is because the third harmonic emitted before the laser focus cancels with that emitted after the focus, owing to geometrical destructive interferences. This cancellation can be circumvented by confining the gaseous region to a hemisphere either before or after the focus,[3–5] or by the use of a narrow gas jet[6] within a region of vacuum.

Net third-harmonic emission can occur, however, in a conventional gas cell if the laser propagation is modified by interacting with the gas, or if the third-harmonic emission deviates from the perturbative law.[7] Malcuit *et al.*[8] showed analytically and experimentally how the nonlinear index of the gaseous medium can distort the phase front of the laser and thus break the phase-interference symmetries to allow third-harmonic emission. Siders *et al.*[9] examined third-harmonic generation in a gas cell under more extreme conditions in which the laser propagation undergoes strong distortions from ionization-produced plasma in the focus. In addition to this symmetry-breaking effect, the perturbative-emission law for the third harmonic is likely no longer applicable as atoms undergo ionization.

Siders *et al.* reported efficiencies as high as 0.01% (one order of magnitude less than in Ref. 1) for third-harmonic conversion of an 800-nm laser pulse in a backfilled chamber of argon gas. Their 80-fs laser pulses had an energy of 1.5 mJ and were focused with *f*/10 optics. When collimating the third-harmonic beam, they observed a strongly blueshifted spatial structure with collimation properties different from that of the rest of the third-harmonic-beam profile. The bandwidth of the blueshifted portion was ~10 nm.

When an intense laser beam is focused in a gaseous medium, the propagation of the beam and hence the nature of the focusing itself is affected. Moreover, for very short (20 fs) laser pulses, there can be complex spatiotemporal interrelations independent of any interaction with atoms.[10] The electronic nonlinearities of the neutral atoms and the temporally and spatially inhomogeneous appearance of free electrons through ionization causes self-phase modulation, defocusing, and blueshifting of the light.[11,12] A number of calculations of the degree of blueshifting are in qualitative agreement with experimental results involving laser pulses of several hundred femtoseconds.[13–15] However, the calculations are performed in one dimension and neglect the interplay of defocusing, an inherently two-dimensional effect. Rae[16,17] performed two-dimensional calculations of a 1-ps pulse interacting with high-density argon gas under tight focusing conditions ($f/5$ optics). He included both the Kerr effect and ionization in the simulation, although based on simple assumptions (e.g., the slowly varying envelope). Even though the peak intensity of the pulse used in the calculation was nominally 10^{16} W/cm^2 in vacuum, his results showed that in an atmosphere of argon, the peak intensity did not exceed a few times 10^{14} W/cm^2 because of defocusing effects.[18]

Rae also produced blueshifting maps of the emerging laser light, where the emerging light was plotted spectrally against a transverse position in the beam, well after the interaction region. The maps showed ring structures of light around the central beam that were blueshifted by as much as $\Delta\omega/\omega \approx 2\%$. These maps are of the same type that we obtained experimentally and present in this paper. Our results cannot be compared directly with the calculations of Rae because we used a much shorter pulse duration and larger f number than those used in his calculations. For our 25-fs pulses, the slowly varying pulse envelope approximation that he used may not be valid. In addition, we produced spectral maps for third-harmonic light, which is outside the framework of his model.

Nibbering *et al.*[19] studied the beam distortions of a 150-fs, 800-nm pulse as it propagated 40 m through air. They observed self focusing to the point that an initially collimated beam converged to reach intensities near 10^{14} 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. They observed rings of light, blueshifted by as much as 200 nm, surrounding the central core at a distance tens of meters after the interaction region. The more blueshifted light occurred at the widest angles.

We present spatially and spectrally resolved measurements of light emerging from the focus of an intense 25-fs laser pulse in gaseous media. The spectral-spatial maps were recorded for various pressures and pulse energies in argon and air. These are, to our knowledge, the first measurements of this type, and they were performed with a laser beam of very good spatial quality. The experiments indicate that the optimal spatial quality of the third-harmonic beam occurs at pressures near ~300 Torr (argon), approximately the same pressure where the highest conversion efficiency takes place. Manifest in these measurements is a broad array of information linked to the interaction of the laser beam with the gas, and this may be used to test the accuracy of future theoretical simulations, which often involve approximating assumptions.

2. EXPERIMENTAL SETUP

The quality of the laser spatial profile is of particular importance in our experiments because small distortions in the laser profile can translate into severe asymmetries in the spatial structure of the radiation emerging from the interaction region. Not only does the emerging radiation depend nonlinearly on the laser beam profile within the interaction region, but this beam profile itself is a nonlinear function involving self focusing and defocusing. When we focus our usual 1-mJ laser pulses[2] into a gas, the fundamental and third-harmonic light beams emerge with complicated and azimuthally asymmetric spatial patterns. This occurs even though the pulses nominally focus to 1.8 times the diffraction limit.

To be able to interpret meaningfully the spatial structures of the emerging radiation, it is necessary to achieve a very clean and well-characterized beam. The degree to which the emerging radiation shows cylindrical symmetry after undergoing nonlinear distortions is a good indication of whether this is achieved. With this aim, we spatially filtered the beam of our 2-TW Ti:sapphire system[20] at the expense of the majority of the pulse energy. This system operates at 10 Hz and can produce up to 50 mJ per pulse so that a large fraction of the energy can be sacrificed while maintaining an energy comparable to the 1-kHz system mentioned above.[2] After strongly filtering the 2-TW beam, we still had available 1 mJ of pulse energy on target, but in a nearly diffraction-limited form. (Some additional damage constraints for mirrors within the filtering setup limited the energy.) Except for these measurements requiring extremely good spatial quality, all our investigations of third-harmonic production in gases have been carried out with the 1-kHz system.[2]

Figure 1 shows a diagram of the experimental setup. After pulse compression, which takes place under vacuum, the 12-mm beam is clipped by a 4-mm hard circular aperture. A concave mirror positioned 72 cm after the aperture focuses the beam to a distance of 1 m where it goes through a 300-μm pinhole. The pinhole blocks all but the central lobe of the Airy pattern induced by the hard aperture. Following the pinhole, the emerging beam travels 252 cm to where a concave mirror of focal length 40 cm refocuses it at a distance of 47.5 cm. One half meter before this focusing mirror, the beam goes through an uncoated 2-mm window made of MgF_2. The window separates the vacuum region wherein the beam is

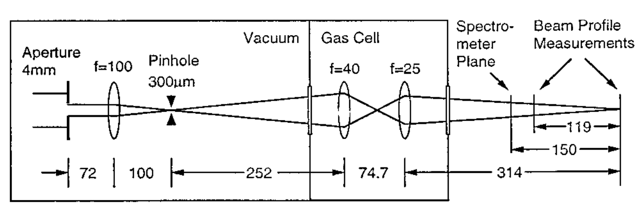

Fig. 1. Experimental setup: The laser beam is apertured and spatially filtered before it is focused inside a gas cell. The emerging light is collected and imaged to a far distance. A spectrometer intersects the beam for spectral analysis as a function of radial position. Near-normal-incidence curved mirrors were used rather than the three lenses depicted for convenience. The dimensions are given in centimeters.

cleaned from the gas-filled region where the interaction takes place. At peak power, the B integral for the window is estimated to be $\pi/20$.

Following the focus in the gas cell at a distance of 27.2 cm, a 1 in. (2.54-cm) concave mirror with focal length 25 cm collects the emerging light and images the focus to a distance of 314 cm. This provides a magnification of the focus of $M = 11.5$. The collecting mirror is coated for high reflectivity of UV light in the range 250–280 nm. The light is reflected from two additional flat mirrors having the same coating (not depicted in Fig. 1). With each reflection the coating attenuates the fundamental light for a total attenuation of $\sim 10^{-3}$, while the third-harmonic light is not attenuated. The light exits the chamber through an uncoated fused-silica window, 2 m before the focal image.

3. LASER PULSE CHARACTERIZATION

Figure 2 shows the measured beam profile taken 119 cm before and at the imaged focus. The beam profiles were recorded electronically by a CCD camera. The images were taken at full power ($\sim$1 mJ) with the gas cell evacuated. The radial contours or line outs shown to the left were extracted from the images displayed to the right. A best-fit Gaussian curve is shown together with the beam profile taken 119 cm before the imaged focus (upper). The curve has a $1/e^2$ radius of $w = 1284\ \mu$m, which is drawn to scale with the image. This indicates an f number of 472 (the picture is taken $\sim$5 Rayleigh ranges away from the imaged focus), which after dividing by the magnification translates into an f number of 41 in the gas cell. Based on this f number, the predicted diffraction-limited waist at the imaged focus is $w_o = 242\ \mu$m, and a Gaussian curve with this waist is plotted together with the measured data, showing very good agreement. A best fit to the data gives an actual waist of $w_o = 258\ \mu$m, 6% over the diffraction-limited case. After dividing by the magnification, the beam waist inside the gas cell is found to be 22 μm.

The pulse duration was measured by autocorrelation to be in the range 25–30 fs. The energy delivered into the gas cell was varied up to $\sim$1 mJ, with an uncertainty in the pulse energy of $\sim$20%. Under vacuum, 1 mJ corresponds to a peak intensity in the focus of $\sim 5 \times 10^{15}$ W/cm^2, or $\sim$20 times above that necessary to ionize argon.[21]

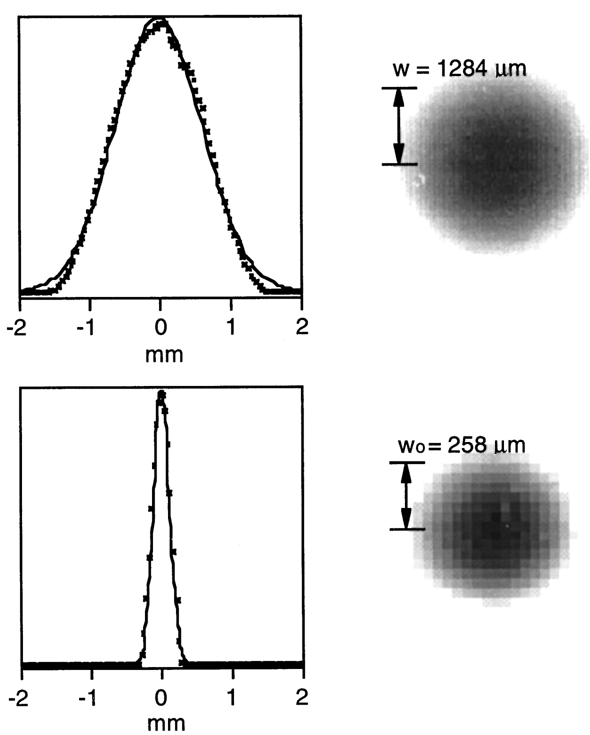

Fig. 2. Radial profiles and corresponding images of the laser beam profile. As indicated in Fig. 1, the beam profile was measured to be 119 cm before (upper) and at the imaged focus (lower). The upper contour data is shown together with a best-fit Gaussian curve. This curve is used to calculate the diffraction-limited curve shown together with the data taken at the imaged focus (lower).

4. SPATIAL-SPECTRAL MEASUREMENTS WITH ARGON

We positioned a spectrometer in the beam exiting the gas cell (see Fig. 1). The slit of the spectrometer sampled a narrow strip of light through the center of the beam. Thus the radial intensity profile of the beam was sampled along the slit, and this light was spectrally resolved in the orthogonal dimension. The spectrometer has no internal imaging in the dimension parallel to the slit, and we estimate the spectrometer CCD screen occurred 150 cm before the imaged focus. While this plane is not the true far field of the focus in the gas cell, it should resemble the far field. Using this setup, we recorded spectral-spatial maps of the emerging light as functions of pulse energy and pressure in the gas cell. We observed the light at wavelengths near the fundamental and the third harmonic.

A. Measurements as Functions of Beam Energy

Figure 3(a) shows a series of maps near the fundamental wavelength (800 nm) captured when laser pulses of various energies are focused in a cell backfilled with 1000 Torr of argon. Figure 3(b) shows a similar series of maps for wavelengths near the third harmonic (267 nm). In all cases, each frame records a single laser shot.

The same spectrometer was used to measure both the fundamental and the third-harmonic pulses, but the pulses were captured individually since the spectrometer grating had to be positioned differently to observe the two wavelength ranges. An absolute uncertainty in wavelength calibration of $\sim$3 nm applies independently to both spectral regions. The spectral resolution is $\sim$1 nm. Faint vertical lines seen on the data in Fig. 3(a) are artifacts of the measurement. The UV-coated mirrors reflect the fundamental light with an efficiency that varies with wavelength by as much as a factor of ten over the range displayed. The response function (including the spectrometer response) was characterized with a white light source, and the result was used to normalize the data shown. We did not characterize the spectral response of our measurements in the UV, but the mirror specifications indicate a flat response over the relevant range.

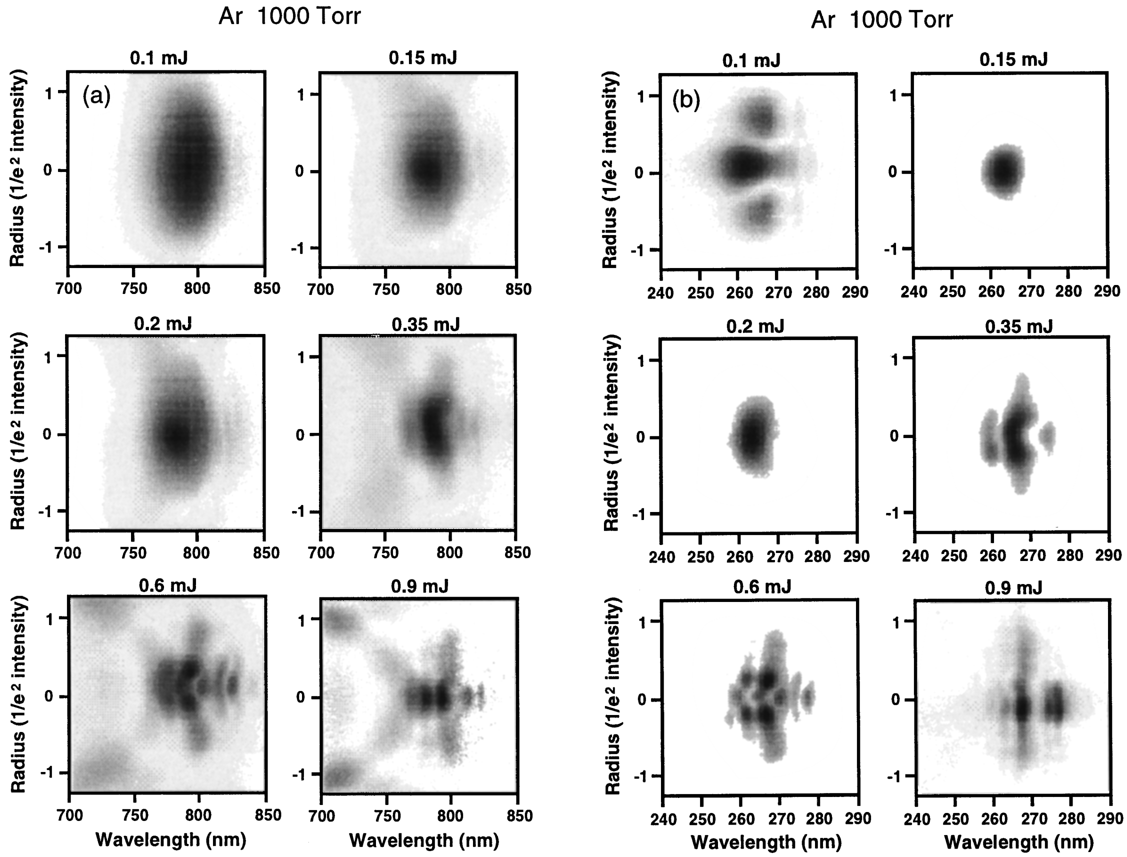

Fig. 3. (a) Fundamental and (b) third-harmonic spectra as functions of beam radial position for various pulse energies focused in 1000 Torr of argon. Position is expressed in units of $1/e^2$-intensity radius of the beam when the gas cell is evacuated. The brightness of the images should not be compared from frame to frame.

Both the fundamental and third-harmonic maps show a general trend of increasing complexity from lower to higher pulse energy. At the lowest pulse energy, 0.1 mJ, the nominal peak intensity in vacuum is $\sim 5 \times 10^{14}$ W/cm^2, a factor of 2 above onset of ionization.[21] Below that energy the third-harmonic signal quickly diminishes, making it difficult to detect with our instrument. All of the images in this paper have been normalized so that on the gray scale the spatial structure can be seen with clarity. Therefore the brightness of the images should not be compared from frame to frame. The peak energy per area on the detector (near center) for the third-harmonic map in the 0.1 mJ case is more than an order of magnitude below that of the 0.15 mJ case. The peak energy per area for the third-harmonic maps continues to increase from frame to frame after that, but only by an additional factor of 5 by the final frame (0.9 mJ), so there is only gradual additional improvement in the efficiency of converting laser light into the third harmonic. At the lowest energy, the third-harmonic emission takes the form of a ring structure surrounding a central spot. As the energy is increased, the central spot becomes dominant to form what looks like a well-behaved beam. At still higher energies, the third-harmonic beam profile becomes much more complex.

The map of the fundamental beam at a pulse energy of 0.1 mJ is very similar to the case of focusing in vacuum. The wide bandwidth (40 nm) is commensurate with the 25-fs pulse duration. However, as the pulse energy is increased, the beam undergoes significant distortions as parts of the fundamental beam are blueshifted. Perhaps the most striking feature within the maps of the fundamental wavelength is a well-defined ring of blueshifted light, which develops at the highest energies.

B. Measurements as Functions of Pressure

Figure 4(a) shows a sequence of maps of the fundamental light obtained for different pressures of argon, all at 0.9 mJ of pulse energy. Figure 4(b) shows a similar sequence of maps showing the third-harmonic light. These sets of maps differ from those of Figs. 3(a) and 3(b) wherein the pulse energy is varied, but there are a number of similarities. For example, at 2.5 Torr and 0.9 mJ the third-harmonic beam shows a ring structure similar to that seen at 1000 Torr and 0.1 mJ. At 100 Torr and 0.9 mJ, the third harmonic forms a beam similar to that seen at 1000 Torr and 0.2 mJ. The fundamental light maps in these cases are also similar.

The peak energy per detector area for the third-harmonic map at 2.5 Torr and 0.9 mJ is the same as that for 1000 Torr and 0.1 mJ. As the pressure increases, the peak energy per area increases by roughly a factor of 5 on each successive frame in Fig. 4(b) up to 100 Torr. The peak energy per area for the map at 100 Torr is a factor of 2 higher than that at 1000 Torr, the highest seen in Fig.

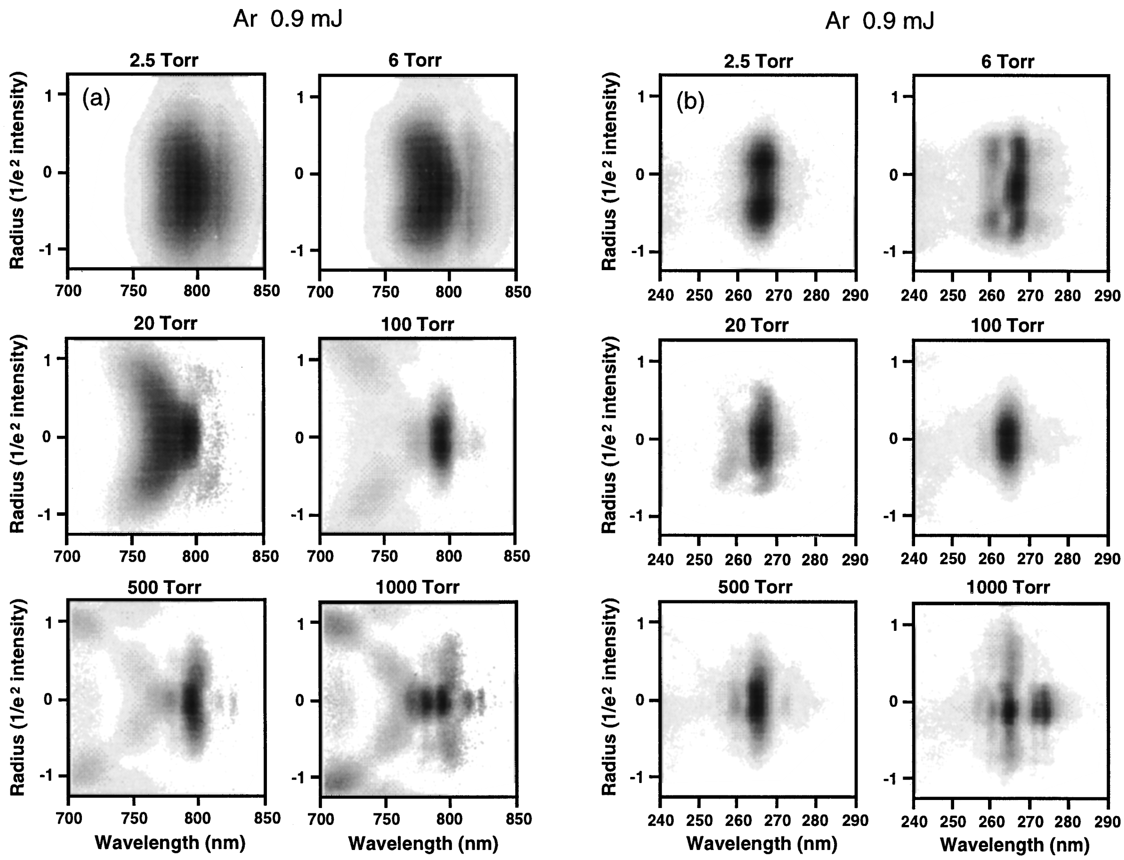

Fig. 4. (a) Fundamental and (b) third-harmonic spectra as functions of beam radial position for a pulse energy of 0.9 mJ focused in various pressures of argon. Position is expressed in units of $1/e^2$-intensity radius of the beam when the gas cell is evacuated. The brightness of the images should not be compared from frame to frame.

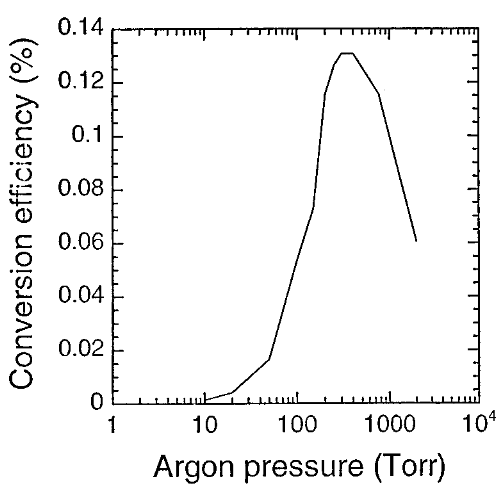

Fig. 5. Conversion efficiency of the third harmonic as a function of argon pressure.

3(b). (Note: The last frame occurs in both figures.) This is in agreement with measurements performed with our 1-kHz laser system[2] focused under similar conditions. Figure 5 shows the conversion efficiency as a function of argon gas pressure. The efficiency of converting the fundamental into the third harmonic was observed to be highest (0.13%) in argon at a pressure in the range of 200–500 Torr. The spectral maps of Fig. 4(b) indicate that this pressure range also yields a well-behaved third-harmonic beam profile.

5. SPATIAL-SPECTRAL MEASUREMENTS WITH AIR

Figure 6 shows a series of maps obtained by focusing various energies in 1 atm of air. Unlike in Figs. 3 and 4, the fundamental light is shown in the left column, while the third-harmonic light is shown in the right column. The results obtained with air have many traits in common with those obtained with argon. For example, the third-harmonic light shows a ring structure for low pulse energy. Also, the map for the fundamental light at 0.9 mJ looks quite similar for the two gases. This suggests that much of the physics involved in the interaction process is species independent. For either argon or air, a plasma spark in the focus approximately 2 mm long was seen with the unaided eye for all of the cases shown. The efficiency in air of converting fundamental light into the third harmonic was generally seen to be about half of that in argon when measured under similar conditions. The fact that the conversion efficiencies are so similar for the two very different gas species is perhaps interesting.

6. DISCUSSION

It appears that the onset of ionization in the focus is linked to the strong emission of the third harmonic. Whether the act of ionization is linked directly with the bulk of the third-harmonic emission or whether ionization plays an indirect role through the breaking of disadvantageous phase-matching symmetries is unknown, but the data suggest that either or both of these is the case. An important aspect of ionization at higher gas densities is the fact that significant energy is required to remove the electrons from the atoms. This is particularly important for our short pulses, which achieve high intensities with minimal pulse energy. Measurements performed with our 1-kHz laser system[2] under similar focusing conditions showed that 20% of the initial beam energy is unaccounted for in the light emerging after the focus. This is similar to the result reported in Ref. 14.

For an undistorted Gaussian beam, the volume wherein the intensity achieves at least one tenth of the peak value can be calculated analytically, and it is found to be $26.2\,w_o^{\,2}z_o$, which for our focusing is 2×10^{-5} cm^3. If we assume for the moment that the laser pulse propagates through gas in the same way as it does through the vacuum, we can calculate the energy necessary to remove at least one electron per argon atom (15.8 eV) inside of this volume. For a pressure of 1000 Torr (3.2×10^{19} cm^{-3}), the required energy is 1.6 mJ, which exceeds the energy of the entire pulse. Of course, this scenario for beam propagation is incorrect because the ionization will strongly affect the focusing. However, the exercise shows that attenuation of the beam owing to ionization is an important element of the problem and should not be neglected. It may also explain at least in part the observed attenuation of the beam energy described above.

An interesting feature of the data at the higher intensities and pressures is the development of a strongly blueshifted ring in the fundamental spectral map. This is particularly evident in the 0.9-mJ case obtained with air as seen in Fig. 5 (lower left). Similar rings have been observed in previous experiments, although under very different conditions (i.e., little or no ionization).[19,22] The numerical simulations of Ref. 17, which included strong ionization, also showed ring structures in the emerging light. The blueshifting presumably arises from rapid ionization in the focus. The wide angle of the ring is perhaps not surprising because the radial density gradient of free electrons in the interaction can defocus the light, or the emission might be thought to originate from a relatively small spot in the focus. However, emission at the blueshifted wavelength is notably absent on the axis, and it is difficult to explain how defocusing or emission from a small spot can cause the far-field pattern to occur in such a well-defined ring structure.

We calculated, by means of Fresnel diffraction, the field pattern necessary to create such a ring structure in the far field, assuming that the light originates from a plane at or near the focus. The ring structure was assumed to have uniform phase, which is not entirely unjustified since the ring maintains itself over a propagation distance of meters. If the light originates from the focal plane, the calculation indicates that it must come from a spot surrounded by rings of alternating phase. The direct occurrence of this specialized pattern in the focus, including phase, seems unlikely. An emission pattern, which already begins in the form of a ring, seems more plausible since a cylinder of plasma is formed in the focus. However, a ring near the focus does not transform into a ring pattern in the far field (assuming an azimuthally

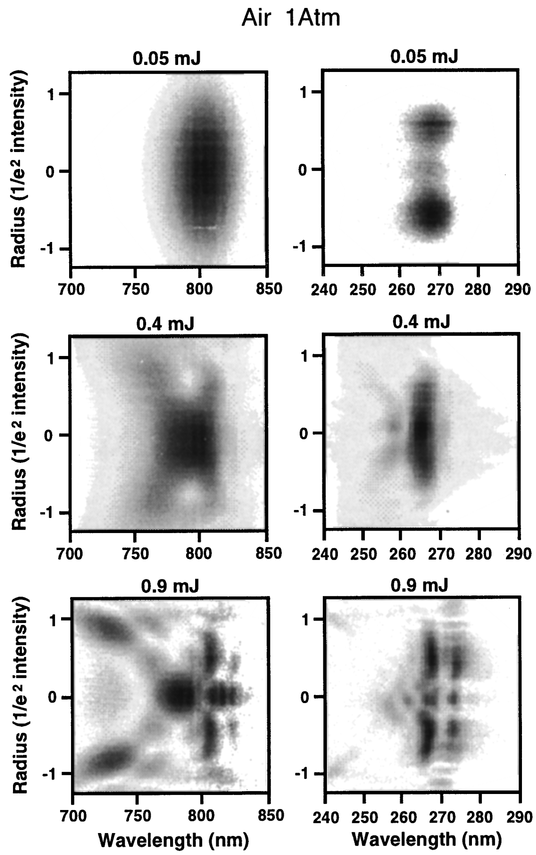

Fig. 6. Fundamental (left column) and third-harmonic (right column) spectra as functions of beam radial position for various pulse energies focused in 1 atm of air. Position is expressed in units of $1/e^2$-intensity radius of the beam when the gas cell is evacuated. The brightness of the images should not be compared from frame to frame.

uniform phase in contrast to a Laguerre–Gaussian laser mode[23]). A ring structure can be preserved, however, if the emission originates from a plane several Rayleigh ranges after or before the focus (or an effective focus). Again, this may seem unlikely, but this would help explain the fact that there is a replica of this ring in the third harmonic, which shows up faintly in Fig. 5 (lower right) with the same divergence angle; if diffraction is involved in forming the ring, one would expect the radial dimensions to differ for the two wavelengths, assuming that the origins of the two rings are connected. The idea of the emission beginning in the form of a ring and continuing to propagate as a ring is consistent with the explanation given in Ref. 17. In any case, the emission likely does not come from a quasi plane but rather from a lengthy volume, and the emission from different axial regions may interfere in a complicated way to form the ring structure in the far field.[19]

We performed additional experiments in argon where the focusing mirror in the gas cell was replaced to create $f/30$ focusing instead of the $f/41$ used to obtain the results presented in this paper. The results for the two cases show many qualitatively similar features, although they differ in quantitative detail. The maps for both the fundamental and the third harmonic obtained with $f/30$ focusing tend to exhibit soft features in comparison with the sharply defined details seen in Figs. 3 and 4, but otherwise they show similar trends. As we tried to go to lower *f* numbers, we found that the azimuthal symmetry of the emitted light began to degrade. This may have been due to subtle distortions in the laser beam profile or to astigmatism introduced by reflecting from the focusing mirror slightly off axis. In any event, the loss of azimuthal symmetry compromises the integrity of the spectral-spatial maps.

7. SUMMARY

We have recorded spectral-spatial maps of radiation emerging from the focus of an intense 25-fs laser pulse in various pressures of argon and air. This survey is done in a range within which it has been determined that $\sim$0.1% of the laser light is converted into the third harmonic, which is of practical importance. We found that the optimal conversion efficiency occurs for the conditions that provide the best spatial and spectral quality of the third-harmonic beam. These measurements are the first of this type and are performed with a laser beam of exceptional spatial quality, considering the intensities involved. In addition, the extreme shortness of the laser-pulse duration provides a unique regime for this type of experiment, where standard slowly varying envelope approximations may not apply. The process of focusing an intense laser into a gas cell is complicated, and these measurements may be useful to compare with numerical simulations, which often employ simplifying approximations. We are working on such calculations and plan to report on them in the future.

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