Tunneling ionization and harmonic generation in krypton gas using a high-intensity, 1-μm, 1-ps laser

S. Augst,[a] D. D. Meyerhofer,[b] C. I. Moore,[a] and J. Peatross[a]

Laboratory for Laser Energetics, University of Rochester
250 East River Road, Rochester, NY 14623-1299

ABSTRACT

Using a high-intensity 1-ps laser we have demonstrated both second and third harmonic production in a jet of krypton gas. The production of even harmonics is a forbidden process in the dipole approximation. Both intensity and pressure scaling for the harmonic strengths were investigated. The laser was operated at 1.053-μm wavelength and ~1-ps pulse length resulting in intensities ranging from 10^{13} to 10^{16} W/cm^2.

1. INTRODUCTION

The study of matter interacting with an intense electromagnetic field has become a topic of interest. Highly nonlinear effects have been seen at intensities above ~10^{13} W/cm^2. Among these effects is harmonic production of the incident laser beam. In atomic gases the dipole approximation predicts the presence of only odd harmonics. We report here the observation of both second and third harmonic production of a 1-μm, 1-ps laser in a jet of krypton gas. The production of third harmonic has been studied extensively; however, to our knowledge second harmonic generation (SHG) in a gas jet has not been previously reported. SHG in a cell of xenon gas has been recently[1] observed and the production mechanism for the two cases appears similar.

2. EXPERIMENTAL SETUP

The laser used for these experiments is a Nd:Glass laser which uses the chirped pulse amplification and compression (CPAC) technique to obtain high-intensity laser pulses and has been described in detail elsewhere.[2] Briefly, a 100-Mhz train of ~40 ps pulses is generated by a Nd:YLF oscillator. These pulses are injected into a single-mode optical fiber where the pulses are temporally stretched and frequency chirped due to group-velocity dispersion and self-phase modulation. The pulses are further stretched by a pair of anti-parallel expansion gratings[3] to a pulse length of ~300 ps. A single pulse is selected from the train and injected into a regenerative amplifier where the energy is increased to ~1 mJ. This pulse is further amplified with two additional Nd:phosphate-glass amplifiers to a maximum of ~100 mJ where the energy is limited by the damage threshold of the compression gratings. For the experiments described here, the pulse is then recompressed with a double-pass through a 1700 l/mm grating pair to a pulse length of ~1 ps. The laser is operated at the fundamental wavelength of 1.053 μm, with a nominal pulse width of 1 ps and a bandwidth of ~20 Å. Using f/5 optics (20-cm focal length lens) intensities ranging from 10^{13} to 10^{16} W/cm^2 were obtained.

The focusing lens is mounted inside a vacuum chamber where the laser is focused onto the output of a Lasertechnics pulsed gas nozzle. The input window on the chamber is covered with an RG1000 filter to prevent laser flashlamp light and other light from entering the chamber. An output window (BK7) is located along the laser axis to allow measurement of second and third harmonics in the forward direction. Laser harmonics are separated with bandpass filters and then detected with a photomultiplier. Initial experiments showed a significant amount of fluorescing in the filters due to absorption of the fundamental IR wavelength. This fluorescence was greatly reduced by placing a 45° dielectric-coated IR mirror before the filters. The second and third harmonics are able to pass through the mirror with a small amount of attenuation, but the fundamental wavelength is attenuated by a factor of ~1000 thereby reducing the intensity incident on the filters.

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Some background harmonic signal was still present using this setup; however, the signal attributable to "other effects" such as harmonic generation in windows and filters was approximately a factor of 100 lower in intensity than the signal from the laser-gas interaction. These residual effects have been subtracted from the data shown in the figures. This was accomplished by performing intensity scans both with and without the gas jet firing. The signal present when the jet was not firing is clearly due to effects other than harmonic generation in the krypton and was therefore subtracted from the signal present when the gas jet was firing. The remaining signal should be solely due to harmonic generation in the krypton gas jet.

A calibration of the gas pressure at the laser focus was performed by measuring the amount of visible light at 45° from the direction of laser propagation. The backing pressure of the jet was varied and the scattered signal measured. The entire chamber was then backfilled to various pressures which produced similar signal levels. The spatial extent of the gas is clearly less in the case of the gas jet which means there are fewer atoms from which the light can scatter; however, most of the scattered light will come from the region of highest intensity so the back-filled chamber should give only a slightly larger signal than the gas jet at equal pressures. We estimate the output pressure of the gas jet at the laser focus to be ~1–2 T (particle density = 3.5–7 x 10^{16} cm^{-3}).

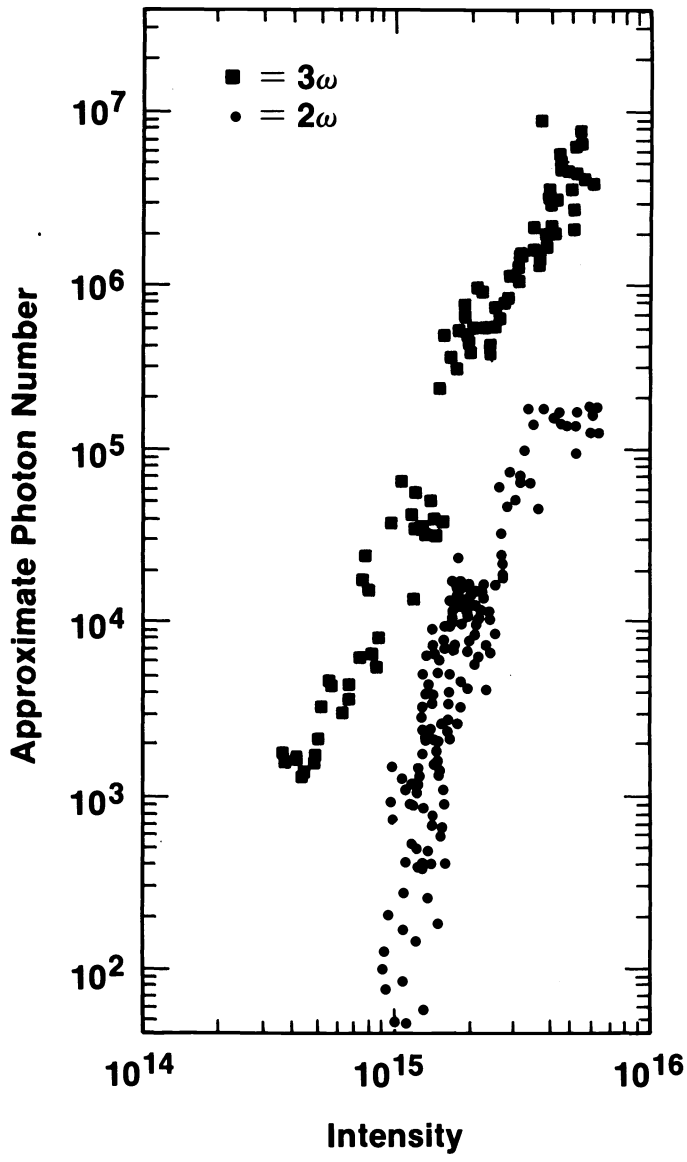
3. HARMONIC GENERATION

It is well known that the production of even harmonics from a laser focused through an isotropic medium is forbidden in the dipole approximation.[4] Other mechanisms for SHG in atomic vapors have been suggested by Bethune.[5] Figure 1 plots our measured harmonic signals in the forward direction for second and third harmonic versus laser intensity. Light at the wavelength of the second harmonic was also seen at 45° from the direction of laser propagation. The temporal duration of this light was 100's of nanoseconds which is characteristic of light from recombination; also, the signal strength was about a factor of 50 less than the second harmonic seen in the forward direction. The duration of the harmonic signal in the forward direction was less than 5 ns, limited by the response time of the photomultiplier.

The production of ions as a function of laser intensity has also been measured under similar conditions and described in detail elsewhere.[6,7] The ion production curves for krypton are shown in Fig. 2. The solid lines are theoretical predictions and will be described in the next section. A comparison of the harmonic signals to the ion signals reveals that second- and third-harmonic production and ion production all commence at approximately the same intensity. It appears that the harmonic production occurs at a slightly higher intensity than the ion production which suggests that an ionized medium may be required to produce both second and third harmonic. There remains a factor of two uncertainty between the ion and harmonic intensity scales since the laser was rebuilt after the ion experiments were completed. The modifications to the laser resulted in slightly different temporal and spatial profiles for the laser beam. We are currently preparing to remeasure the intensities for ion production to determine under what plasma conditions harmonic production occurs.

The second harmonic signal is characterized by a steep slope for low intensity and a less steep slope at higher intensities. Since the steep slope occurs just above the intensity where ionization of krypton begins, the harmonic production is probably a result of plasma effects. The most probable effect is SHG due to a static electric field at the laser focus. Electrons liberated at the focus have some kinetic energy due to "above threshold ionization" (ATI) effects. Additional energy is imparted to them from the ponderomotive potential which for us is 500 eV at 5 x 10^{15} W/cm^2. Due to the length of the laser pulse, the electrons can acquire only a fraction of the ponderomotive energy. We have measured electron energies approaching 100 eV. Since the ions are much more massive, they leave the focal volume more slowly than the electrons, thereby creating a charge separation consisting of positive ions surrounded by a cylinder of negative electrons. A radial electric field results which breaks the symmetry of the medium, thus allowing SHG to occur through a process which is effectively 4-wave mixing between two laser photons and the static electric field.

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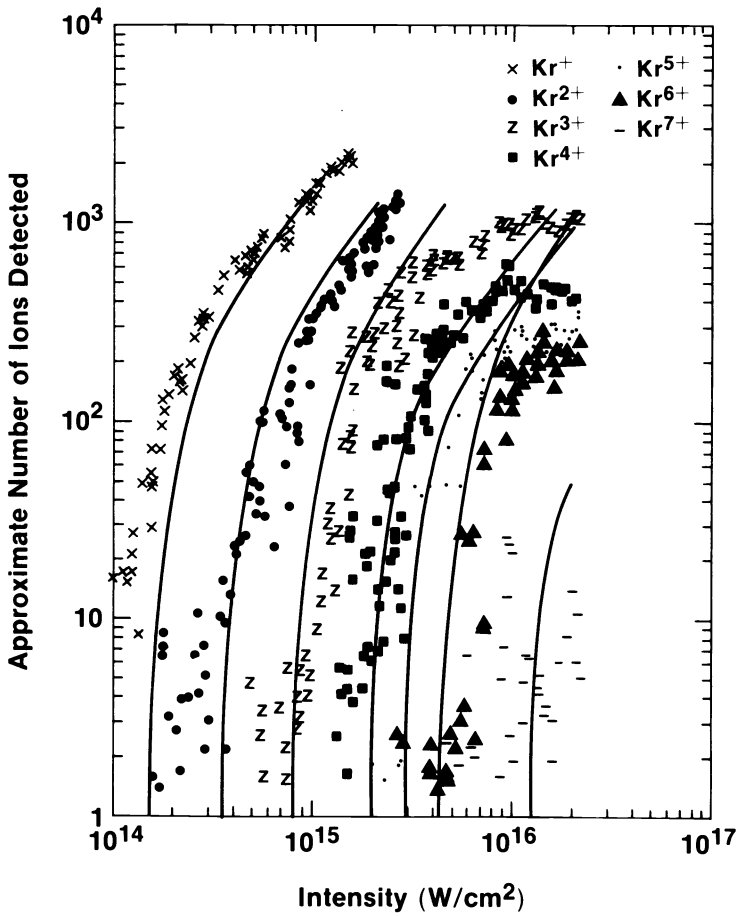


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Fig. 1. Second and third harmonic production in a krypton gas jet as a function of laser intensity. The gas pressure is ~1.3 T.

The experimental conversion efficiency for second harmonic (using $I = 10^{15}$ W/cm^2) was measured as $\eta_2 = 1 \times 10^{-14}$ with a factor of three uncertainty. The magnitude of the static electric field can be estimated as $\varepsilon_o = \Phi/ew_o$ where Φ is the ponderomotive potential, e is the electron charge, and w_o is the beam waist radius at focus. At an intensity of 10^{15} W/cm^2, we find a value for the static electric field of $\varepsilon_o = 8 \times 10^4$ V/cm. This is used to calculate the expected theoretical conversion efficiency[1] $\eta_2 = 9\,\pi^3 k_1^4\,[\chi^{(3)}]^2\, I\, \varepsilon_o^2\, w_o^4/c$, where k_1 is the fundamental wave number and $\chi^{(3)}$ is the nonlinear susceptibility given by $\chi^{(3)} = N\gamma$, where $N = 5 \times 10^{16}$ cm^{-3} and $\gamma = 3.8 \times 10^{-37}$ esu.[8] We find a theoretical value of $\eta_2 = 7 \times 10^{-14}$, which is within order-of-magnitude agreement with our experimental result. The discrepancy may be due to uncertainties in the ion density.

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Fig. 2. Krypton ion production rate as a function of laser intensity. The theoretical curves are from BSI theory and are described in Sec. 4. The gas pressure is 5×10^{-6} T.

The third harmonic signal has an intensity dependence of I^3 as expected; however, a comparison to longer pulse length experiments at the same wavelength[9–11] reveals two interesting differences. First, third harmonic production for 30-ps laser pulses is evident at intensities lower than those required to ionize the medium and a saturation of the harmonic signal occurs as the laser intensity is increased past the saturation point of the first ionic charge state. For 1-ps laser pulses, no third harmonic signal is seen until the gas is ionized and as the laser intensity is increased, no saturation is observed. This suggests that the harmonic production mechanism may be different for 1-ps pulses than it is for 30-ps pulses. It is unlikely, however, that the third harmonic production is due entirely to plasma effects since the third harmonic signal is larger than the second harmonic signal. Second, the intensity at which the ions and harmonics are first seen is much lower in the longer pulse length experiments. This intensity shift is not yet fully understood; however, it introduces an interesting feature. For 1-ps pulses, the ionization process is pushed from the multi-photon regime into the tunneling regime. We have demonstrated that all ionic charge states for krypton appear in the tunneling regime[6] defined by $\gamma < 1$ (where $\gamma = (E/2\Phi)^{1/2}$ is the Keldysh intensity parameter;[12] E = ionization potential, and Φ = ponderomotive potential). The shift from the multi-photon regime to the tunneling regime may be responsible for differing harmonic production mechanisms.

We have also measured harmonic signal strength as a function of gas density. Pressure scans for second and third harmonic are shown in Fig. 3. The pressure dependence for the production of third harmonic is $\sim P^2$ as expected,[11] however, the dependence for the second harmonic is $P^{1.2}$. This is in contrast to the P^3 pressure dependence seen in a gas cell[1] which is

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in agreement with Bethune's prediction.[5] It is unlikely that experimental uncertainty can account for this discrepancy. At present we have no explanation for the $P^{1.2}$ dependence.

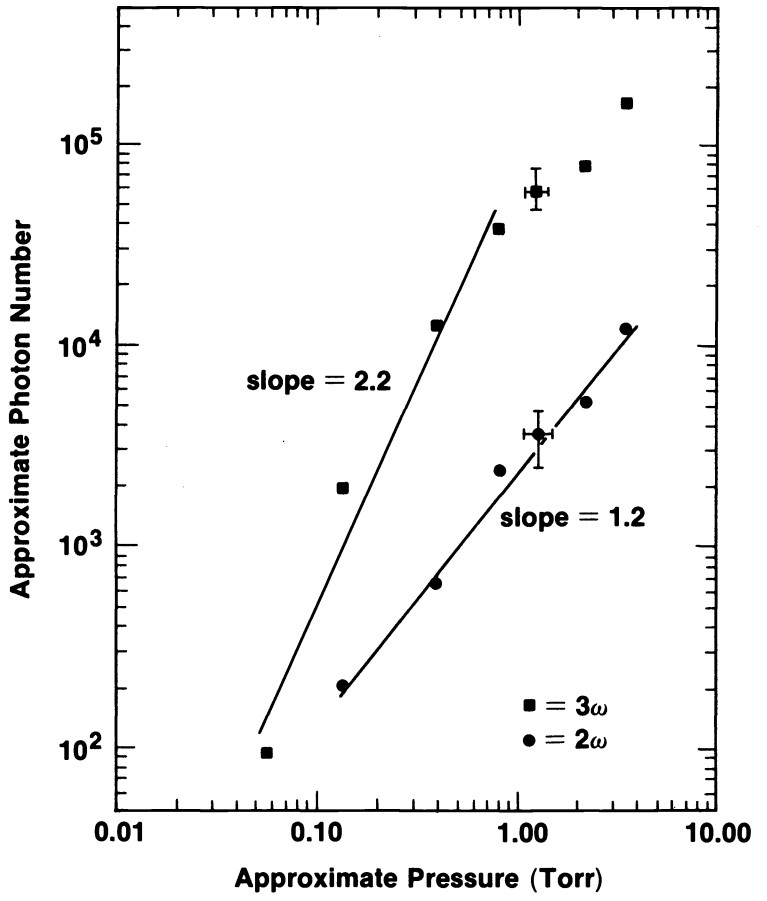


Fig. 3. Second and third harmonic production in a krypton gas jet as a function of gas pressure. The laser intensity is ~4 x 10^{15} W/cm^2.

4. ION SPECTROSCOPY

The solid lines shown over the ion production curves in Fig. 2 are from theoretical calculations. We have called this simple one-dimensional model theory of ionization "Barrier Suppression Ionization Theory" (BSI). BSI begins with the superposition of the Coulomb potential and a quasi-static laser field. The resulting total potential is written as:

$$V(x) = \frac{Ze^2}{x} - e\mathcal{E}x. \tag{1}$$

This potential has a relative maximum located at x_{max} which can easily be found by setting $[\partial V(x)/\partial x]_{x = x_{max}} = 0$ and solving for x_{max}. Equating $V(x_{max})$ to the ionization potential (E) of the atom or ion permits us to find the critical electric field which is necessary to allow the bound electron to escape without tunneling. This critical electric field is:

$$\mathcal{E}^2 = \frac{E^4}{16e^6Z^2} \ . \tag{2}$$

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When the laser electric field equals this critical field strength, the atomic electron can freely escape, and the corresponding laser intensity can be considered to be the appearance intensity:

$$I_{app} = \frac{cE^4}{128\pi e^6 Z^2} \cdot \tag{3}$$

In a purely Coulombic field the ionization potential is uniquely dependent on the ionic charge Z and the principal quantum number n: $E = (m_e Z^2 \alpha^2 c^2)/(2n^2)$. However, for the complex atoms used in our experiments, electron shielding occurs and its effects can be accounted for by using values of E which have been determined independently, either experimentally or by use of more sophisticated atomic structure calculations.

The ion production curves for BSI theory are obtained by integrating the coupled rate equations for the number of ions in each charge state. The spatial and temporal dependence of the intensity, and consequently of the ionization rates, is also accounted for in the calculation. An integration is performed by finding the volume of each iso-intensity shell and then propagating a laser pulse through that volume. The number of particles in each shell is conserved to account for saturation effects. The intensity distribution is modeled as temporally Lorentzian and spatially Gaussian.[13] The ion spectra calculation assumes that the ionization probability is unity for $I \geq I_{app}$ and zero for $I < I_{app}$. The resulting BSI theory curves shown in Fig. 2 use a one-parameter fit, this being a shift in the vertical scale which simply accounts for spectrometer efficiency. There has been no shift along the intensity scale for the theoretical curves, thus these are effectively zero-parameter fits to the ionization . Similar graphs have been made for helium, neon, argon, and xenon. In all cases, the agreement is excellent.

5. SUMMARY

We have seen second and third harmonic generation of a 1-μm, 1-ps laser in a jet of krypton gas. Under these laser conditions, the production mechanism for both second and third harmonic requires the presence of an ionized medium. Third harmonic results differ in several ways from longer pulse experiments at the same laser wavelength, but second harmonic results are in basic agreement with experiments done in a gas cell.

Current experiments include: (1) a characterization of the plasma conditions under which harmonic generation occurs, (2) an investigation of harmonic and ion production for laser pulse lengths between 1 ps and 30 ps, and (3) an investigation of higher order harmonic production.

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(b) Also Department of Mechanical Engineering , University of Rochester.
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