High-order Harmonic-generation In Laser-produced Ions Using A Near-infrared Laser

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High-order harmonic generation in laser-produced ions using a near-infrared laser

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We describe experimental studies of high-order harmonic generation in rare-gas-like Na^+ and K^+ ions. The ions were created in a laser-produced plasma and the harmonics were generated with a short-pulse, high-power, titanium-sapphire laser operating at 794 nm. The highest harmonic order observed was the 27th (29.4 nm), while much higher harmonic orders were observed in a jet of neutral Ne gas under similar conditions. Pronounced ring structures were observed in the harmonic far-field distribution and these structures varied with focusing condition. The cutoff in the harmonic spectra at high orders may be due to ionization-induced defocusing of the fundamental laser beam. Calculations illustrating how this effect reduces the peak intensity obtained in the plasma compared with the intensity obtained in the Ne gas are presented.

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I. INTRODUCTION

High-order harmonic generation using high-power lasers has been found to be a very promising way of generating ultrashort pulses of intense, coherent radiation in the xuv and the soft-x-ray range. Using rare-gas atoms as the nonlinear medium, harmonic orders as high as 135 [1] and wavelengths as short as 7.4 nm [2] have been reported using short-pulse lasers in the infrared and near infrared. The most characteristic behavior in the distribution of the intensities of the harmonics is a plateau where the intensities of successive orders are roughly the same, followed by a rather sharp cutoff beyond which the harmonic intensities fall off rapidly. Photons with energies far exceeding the atomic binding energy are produced.

For the high harmonics, the cutoff energy, i.e., the photon energy of the highest harmonic in the plateau, has been found to increase linearly with the laser intensity [3]. This increase, however, does not continue in an unlimited way. As a specific intensity is reached—the saturation intensity I_{sat}—considerable ionization occurs and the harmonic generation saturates, mainly due to the depletion of the nonlinear medium. The saturation intensity is species dependent; therefore the maximum extent of the plateau will also depend on the species used as the nonlinear medium. It is well known that among the noble gases, the extent of the plateau increases with the ionization potential (i.e., higher harmonics can be generated in He and Ne than in Kr and Xe). On the other hand, for a harmonic of a given order within the plateau, the yield of photons decreases with the ionization potential.

Numerical calculations, valid in the strong-field, low-frequency regime, have shown that the maximum energy at the end of the plateau in the single-atom response is well approximated by the simple formula I_p+3U_p, where I_p is the ionization energy and $U_p=9.33\times10^{-14}I\lambda^2$ is the ponderomotive energy (in eV) of the electron in the laser field with intensity I (in W/cm^2) and wavelength λ (in μm) [4]. A rather simple picture, which gives some insight into the physical meaning of this formula, can be found in Refs. [5,6]. The effects behind this formula can be understood in terms of tunneling ionization, followed by acceleration in a classical field and recombination with the ionic core. The linear intensity dependence observed experimentally [3] had a somewhat smaller constant of proportionality than the single-atom response ($\approx 2.4U_p$ instead of $3U_p$). This difference was explained as propagation effects in the tight-focusing regime, applicable to the high-order harmonics [7].

The ponderomotive energy is proportional to the square of the laser wavelength. In a given nonlinear medium, a low-frequency laser can therefore generate harmonic radiation with shorter wavelengths than a high-frequency laser, even if much higher harmonic orders are required.

Theoretical studies [4,8] indicate that by using various ions, e.g., He^+, very high harmonics, above 400 eV, should be possible using low-frequency, high-intensity lasers. Rare-gas-like ions from singly ionized alkali metals also have very high ionization potentials and saturation intensities. The ionization potentials of Li^+, Na^+, and K^+ are 76.5, 47.3, and 31.6 eV, respectively, while the corresponding values for the rare-gas atoms He, Ne, and Ar are 24.6, 21.6, and 15.8 eV, respectively. The saturation intensities for ionization can be estimated using, e.g., expressions for barrier suppression [9] or tunneling ionization [10]. Using the scaling rules in [9], one finds that an increase in ionization potential of about a factor of 2 between a neutral atom and a singly ionized ion, such as that between Ne and Na^+, results in an increase in the corresponding saturation intensity of about a factor of 4. From the I_p+3U_p formula, and the fact that the maximum value of U_p is proportional to I_{sat}, one can expect to be able to generate much higher harmonic orders in these ions compared with those in the corresponding

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rare-gas atoms.

Using gas targets, a large number of studies have been reported, using different laser wavelengths, pulse duration and focusing geometry. Only recently has the possibility of using rare-gas-like ions started to be investigated experimentally. The first reports are by Kubodera *et al.* [11]. These authors used a nanosecond KrF laser to ablate plasmas from various metal targets and a subpicosecond KrF laser to generate harmonics in these expanding plasmas. They observed, e.g., harmonics up to the 19th (13.1 nm) in a sodium plasma and up to the 13th (19.1 nm) in a potassium plasma.

The purpose of the present work was to study the extent of the harmonic spectrum obtained with a high-intensity, near-infrared laser in a laser-produced alkali-metal plasma and to investigate how the harmonic efficiency depends on various experimental parameters and how these can be optimized.

After this introduction, we will describe, in Sec. II, the experimental arrangement and how various experimental conditions were optimized. The results are presented in Sec. III and discussed in Sec. IV. Finally, in Sec. V we summarize and present the conclusions drawn from this work.

II. EXPERIMENTAL ARRANGEMENT

The schematic outline of our experimental setup is presented in Fig. 1. The laser beam (diameter 50 mm) from a high-power, 150-fs, titanium-sapphire (Ti:S) laser, operating at 794 nm [12], was focused by a plano-convex lens ($f=1.0$ m) into a vacuum chamber. At the focus it interacted with an expanding plasma generated by a Nd:YAG laser operating at 1064 nm, focused onto a solid alkali-metal target by an $f/30$ cylindrical lens. The line focus was oriented parallel to the propagation axis of the Ti:S laser. The length of the interaction region, i.e., the plasma, was about 10 mm and the density of atoms and ions was estimated to be of the order of 10^{17} cm^{-3} in the interaction region. The harmonic radiation generated and the laser beam propagate in the same direction after the interaction region and a grazing incidence xuv spectrometer was used to separate the laser radiation and the harmonics of different orders. In order to obtain a high collection efficiency, it would be advantageous to place the entrance slit close to the interaction region. However, at this point the diameter of the diverging laser beam is still small, the intensity is very high, and there is a serious risk of laser-plasma generation on the entrance slit or optical damage to the grating. The sputtering of debris from the laser-produced plasma constitutes a further reason for placing the spectrometer at a greater distance. We used a 100-μm-wide (10-mm-high) entrance slit on the spectrometer and placed it at a distance of 0.9 m from the focus. This means that we only recorded harmonics emitted within a 0.1-mrad-wide cone. However, by placing the entire spectrometer on a precision translation stage, allowing translation in the direction perpendicular to the optical axis, we could use this small acceptance angle to scan the angular distribution of the harmonics.

Finally, the harmonics were detected using an electron multiplier tube (EMT) and recorded with a boxcar integrator and a computer-based data acquisition system. Using the boxcar integrator, we integrated only the signal that arrived within a given time window (chosen to be 60 ns wide to match the temporal response of the electron multiplier tube) centered around the arrival time of the Ti:S laser pulse. This was essential in order to suppress the light emitted from the laser-produced plasma. The choice of delay time between the two lasers will be discussed below.

Each alkali-metal target was fabricated by rolling out a

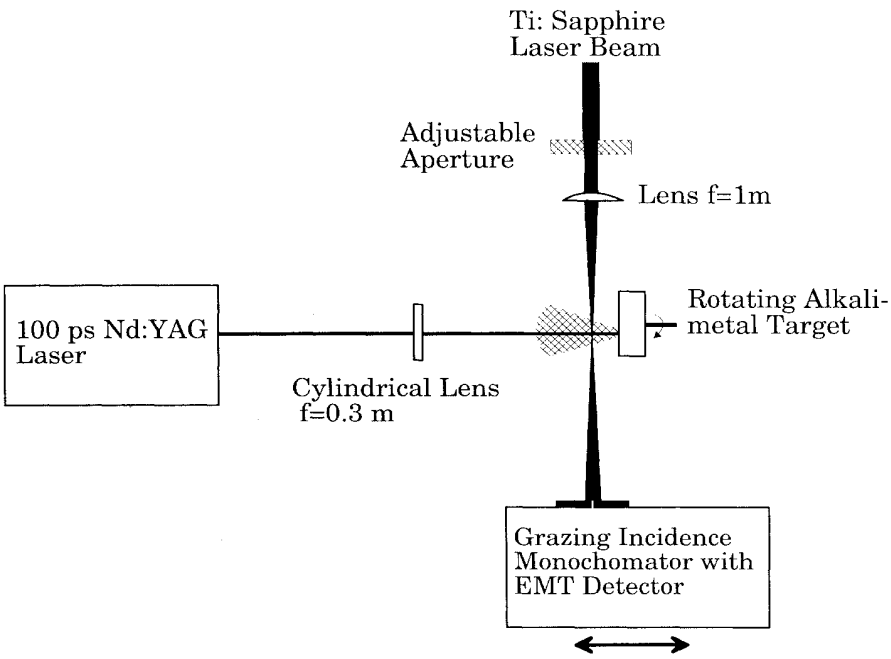

FIG. 1. Experimental setup for the generation and the detection of high-order harmonics in a laser-produced alkali-metal plasma.

solid piece of metal to a uniform sheet of 1 mm thickness. This sheet was then pressed onto a thick polyethylene substrate of 55 mm diameter mounted on a rotating shaft. The surface of the plastic substrate was made sufficiently rough to make the sheet of metal stick mechanically. The rolling out and the mounting of the target were performed in air, but due to the reactivity of the alkali metals, this had to be done quickly to avoid excessive layers of oxide forming before the targets were placed under vacuum. The thin oxide layer inevitably formed was ablated away during the first few minutes of irradiation with the plasma-generating laser.

Two different Nd:YAG lasers (where YAG denotes yttrium aluminum garnet) with comparable pulse energies were available for the plasma generation: one with a pulse duration of about 10 ns, the other, being a mode-locked laser, with about 0.1 ns pulse duration [12]. The harmonic yield as a function of pulse duration was investigated by switching between these two lasers and in each case optimizing the delay between the plasma generation and the high-power pulse, keeping other parameters constant. We found no dramatic difference in the yield, but possibly a slight increase in signal when going from 10-ns pulses to 0.1-ns pulses. However, when going to the shorter pulses, we noticed a significant decrease in the amount of continuum plasma light and in the amount of sputtered debris in the target chamber and on the detection optics. The plasma light was not a major problem since we used a gated detection system. The sputtering of debris, on the other hand, was one of our major concerns in this experiment. We therefore chose to use the mode-locked laser with 0.1 ns pulse duration for plasma generation during the rest of the study.

In order to optimize the yield of the harmonics, the dependency of a number of parameters was explored. One such parameter was the energy of the plasma-producing laser pulses. We found that, for constant focusing conditions, the harmonic intensity increased approximately exponentially with the energy in the plasma-producing pulses up to about 55 mJ. Above this energy the signal did not increase any further upon increasing the pulse energy. Increasing the energy further only increased the stray plasma light and sputtering of debris in the vacuum chamber and onto the detection optics. The pulse energy was therefore kept constant at about 55 mJ/pulse during the rest of the study.

The distance between the target and the focused Ti:S laser beam and the time delay between the plasma-producing Nd:YAG laser and the Ti:S laser are two other parameters that are important in the optimization of the harmonic signal. The harmonic signal was found to decrease approximately exponentially with the distance from the target. This is illustrated in Fig. 2, where the signal of the 17th harmonic is plotted as a function of the distance, together with an exponential curve fitted to the data. The $1/e$ decay length of this fitted curve is 0.46 mm. Most of the data presented below were obtained at a distance of 0.8 mm from the target. Choosing a shorter distance made the experiment unnecessarily sensitive to small variations in the surface of the rotating target.

Maintaining the distance from the target constant, we investigated how the harmonic signal varied with the delay between the two lasers. We found that the harmonic yield exhibited a maximum when the plasma-producing laser pulse arrived at the target about 200 ns before the high-intensity Ti:S laser pulse. (When using the 10-ns laser for the plasma generation, about a three times longer delay was required to maximize the harmonic signal.) The interpretation of this result is not completely clear. The signal could be expected to increase monotonically with the local density of the expanding plasma. However, effects such as phase matching, reabsorption of the generated radiation, and defocusing of the laser beam also depend on the local density. One can not simply assume, therefore, that the time delay corresponding to the maximum harmonic signal also corresponds to the maximum local density in the plasma. In the rest of the study we kept the delay between the two lasers fixed at 200 ns.

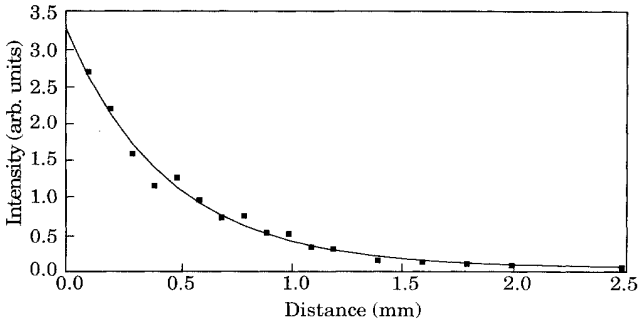

FIG. 2. The intensity of the 17th harmonic in Na^+ as a function of the distance between the surface of the solid target and the focus of the Ti:S laser. An exponential curve, fitted to the data, is included.

III. EXPERIMENTAL RESULTS

A. Harmonic spectra in Na and K plasmas

The harmonic spectra obtained in sodium and potassium are shown in Figs. 3(a) and 3(b), respectively. Only the target material was changed between the recordings of the two spectra; all the other experimental parameters were kept constant. The energy in the plasma-producing laser pulses, the time delay between the two lasers, and the distance from the target were all kept at their optimized values, as described in the preceding section. The spectra are very similar, showing a very rapid decrease with harmonic order. In both spectra the highest harmonic orders observed were about the 27th. In order to be able to make a comparison with the more well-known spectra from rare gases, we replaced the rotating solid target with a pulsed gas nozzle, keeping other parameters such as laser-pulse energy, focusing, and the detector sensitivity constant. Part of the Ne spectrum obtained in this way is shown in the inset of Fig. 3(a). The plateau in this spectrum extended to at least the 59th harmonic. The spectra have not been corrected for spectral efficiencies of the monochromator and the detector. However, the long plateau observed in the Ne spectrum

indicates a relatively flat response over a large spectral region and in particular over the region covered by the Na^+ and K^+ spectra.

B. Focusing conditions

The importance of the focusing geometry for the macroscopic harmonic efficiency is well known. A useful quantity that characterizes the focus is the confocal parameter $b = 2\pi w_0^2/\lambda$, where w_0 denotes the beam radius at the focus. For a Gaussian beam, b is equal to twice the distance on the propagation axis over which the beam cross-sectional area increases by a factor of 2. It has been found, using rare gases as the nonlinear medium [13,14], that in the tight focusing limit, the measured number of photons is proportional to b^3. A simple way to vary the confocal parameter is by a variable aperture placed in the laser beam, before the focusing lens. By decreasing (radially truncating) the diameter of the laser beam, the beam waist at the focus increases and hence also the confocal parameter. However, increasing the focal cross section implies a corresponding reduction in intensity.

In an attempt to find the best compromise between the intensity and the focal cross section, we recorded the harmonic signal as a function of the diameter of the hard aperture placed in the laser beam before the focusing lens. As illustrated in Fig. 4 for the 17th harmonic, a series of three maxima, separated by distinct minima, was obtained as a function of aperture diameter. This is quite different from the b^3 relationship observed with rare gases as the nonlinear medium, leading to only one maximum. Repeating the measurement, but instead observing the radiation at a small angle (≤ 2 mrad) from the optical axis, we found that the first maximum always appeared at an aperture diameter of 15 mm. However, the second and third maxima, corresponding to 28 and 38 mm aperture diameters, respectively, in Fig. 4, where shifted and occurred at successively larger aperture diameters as the distance from the optical axis was increased.

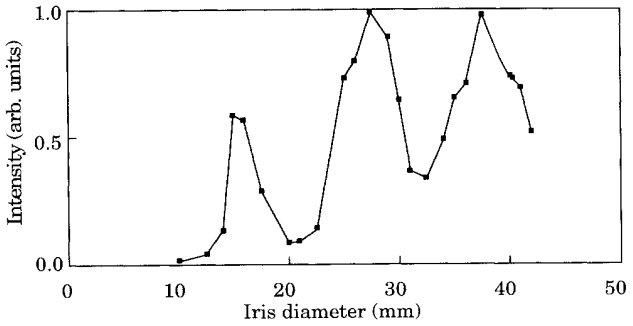

FIG. 4. Plot of the 17th harmonic in Na^+ as a function of the diameter of the aperture placed before the focusing lens. The data were obtained with the spectrometer slit placed on the optical axis.

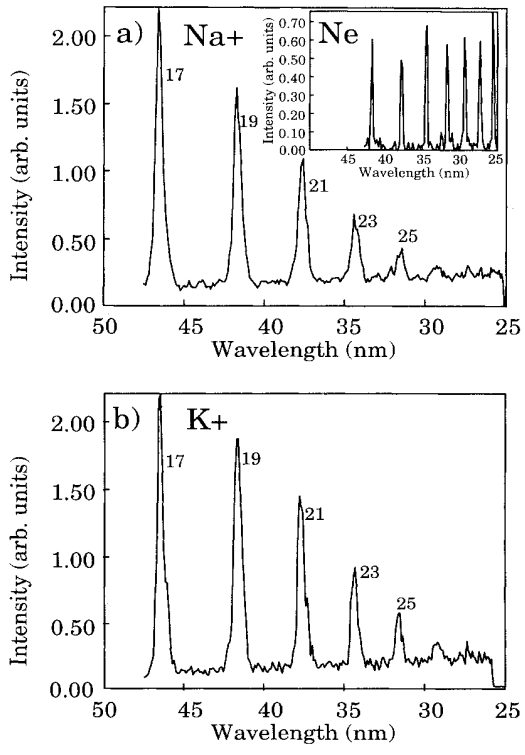

FIG. 3. Harmonic spectra obtained (a) in Na^+ and (b) in K^+. The inset in (a) is part of a spectrum obtained under similar conditions, but with the laser-produced plasma replaced with a pulsed nozzle producing a jet of neutral Ne atoms. The intensity is given in arbitrary units, different for each spectrum.

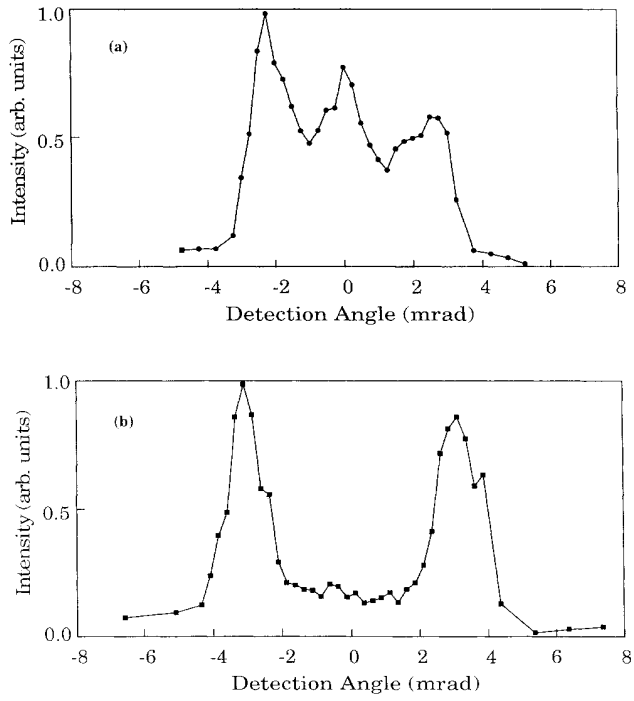

FIG. 5. Illustration of the angular distribution of the 17th harmonic in Na^+, recorded by translating the entire spectrometer in the direction perpendicular to the optical axis of the experiment. The results were obtained with (a) a 15-mm and (b) a 20-mm aperture, corresponding to the first maximum and minimum in Fig. 4, respectively.

C. Angular distributions

In order to understand this series of maxima and minima in the harmonic intensity versus the focusing condition, we studied the angular distribution of the same harmonic, but with different aperture diameters. Choosing an aperture diameter of 15 mm, corresponding to the first maximum in Fig. 4, the angular structure shown in Fig. 5(a) was observed. This structure shows a central peak in the direction of the optical axis, the direction of observation used when studying the harmonic signal versus the aperture diameter, and symmetric peaks further out. We attribute these peaks to a ring structure in the far-field distribution of the harmonics. On the other hand, choosing an aperture diameter of 20 mm, corresponding to the first minimum in Fig. 4, gave the result illustrated in Fig. 5(b). In this case there is also a ring structure, but without the central peak. Similar structures were also observed for other harmonic orders. The harmonic far-field distribution hence exhibits distinct angular structures and these structures vary with focusing conditions.

IV. DISCUSSION OF THE RESULTS

These experimental results raise a series of questions. First, are the observed harmonics due to neutral atoms or to ions? Second, why do we obtain a rapid decrease with harmonic order at relatively low orders in the alkali-metal spectra compared with the spectrum obtained in a Ne gas jet? Is it due to propagation effects, i.e., phase matching, in the plasma or does it reflect the single-atom (-ion) response, related to the intensity in the interaction region?

A. Contribution from neutral atoms

The ionization potential of neutral Na is only 5.1 eV. Generation of, say, the 21st harmonic (33 eV) therefore implies a photon energy 28 eV higher than the ionization potential. Applying the simple formula I_p+3U_p for the plateau cutoff energy, one could estimate that the ponderomotive potential U_p required would be 9.3 eV. This corresponds to an intensity of 1.6×10^{14} W/cm^2 and we would be in the domain of tunneling ionization required for the I_p+3U_p formula to be valid. However, the saturation intensity for ionization of neutral Na, using 150-fs pulses at 794 nm, is only about 10^{12} W/cm^2 [19]. Thus the atom will be ionized through mulitphoton ionization during the rise time of the laser pulse if higher intensities are applied. We therefore conclude that the high-order harmonics we observed were generated in the rare-gas-like ions and not in the neutral atoms. Following the discussion in the Introduction, one could therefore expect to generate higher harmonic orders in these alkali-metal plasmas than in neutral Ne, contradictory to our observations.

B. Phase matching in the presence of free electrons

The creation of a macroscopic field requires that there be proper phase matching between the induced and the driving fields [15]. There are essentially two reasons for the phases not to be matched. First, the dispersion in the nonlinear medium makes waves at different frequencies travel at different speeds in the medium and therefore become out of phase with each other. The length in the medium over which the harmonic field and the polarization field are in phase, before dispersion brings them out of phase and destructive interference begins, is given by the "coherence length" $L_{\rm coh}^{\rm disp}$. The atomic density in our experiment was low, so the dispersion from neutral atoms and ions was small. However, the presence of free electrons due to ionization of the medium has a non-negligible effect on the refractive index. This can introduce a substantial phase mismatch between the generated harmonics and the driving polarization [15]. This phase mismatch is proportional to the density of free electrons to the wavelength of the laser (this is why the free-electron dispersion is less important when using, e.g., excimer lasers in the uv) and to the harmonic order q (for high orders). With our laser wavelength and assuming an electron density of about 10^{17} cm^{-3}, $L_{\rm coh}^{\rm disp}$ is approximately 1 mm for the 13th harmonic and decreases with harmonic order as $1/q$.

The second reason for the phases not to be matched is connected with the unavoidable phase shift of π, occurring in a light wave upon passing through a focus [16]. Using a loosely focused laser beam and a finite nonlinear medium, only a fraction of this phase shift occurs in the medium. However, the difference in phase between the driving field and the generated field will be $(q-1)$ times as large for a harmonic of order q. For sufficiently high harmonic orders, therefore, the phase lag will always reach π after a certain length inside the medium, resulting in destructive interference. Close to the focus, this length, the coherence length due to focusing $L_{\rm coh}^{\rm foc}$, is proportional to the confocal parameter and to $1/(q-1)$ [14]. In our study we varied the diameter of the fundamental laser beam, and hence the confocal parameter, being inversely proportional to the square of the beam diameter. Using a beam aperture diameter of, say, 16 mm together with our $f=1m$ focusing lens, the confocal parameter is about 7 mm and $L_{\rm coh}^{\rm foc}$ for the 13th harmonic becomes approximately 1 mm, decreasing with the harmonic order as $1/(q-1)$.

A complete picture of the macroscopic propagation effects is very complex. However, the amplitude of the harmonic field at the point of observation, outside the medium, is closely related to the length in the medium over which the wave can build up coherently [14]. This length can be estimated as the shortest of the two coherence lengths described above. This leads to two important conclusions. First, when working with rare gases, it is usually advantageous to use loose focusing, as $L_{\rm coh}^{\rm foc}$ is proportional to the confocal parameter. However, in a case like ours, the free-electron dispersion will make $L_{\rm coh}^{\rm disp}$ shorter than $L_{\rm coh}^{\rm foc}$ and increasing the confocal parameter will have much less effect. The observed signal intensity is hence reduced by the presence of the free electrons. Second, both the coherence length due to focusing and the coherence length due to dispersion scale with harmonic order approximately as $1/q$. The phase-matching effect on the observed harmonic intensity will therefore

scale with harmonic order approximately as $1/q^2$. However, the intensities of the observed harmonics [Figs. 3(a) and 3(b)] decrease with harmonic order considerably faster than as $1/q^2$. We therefore conclude that phase matching is not the reason for the rapid decrease in the observed spectra and that the explanation must be the single-atom response.

C. Single-atom response

In the regime of tunneling ionization, the extent of the plateau in the single-atom response can be estimated from the I_p+3U_p rule. The intensity required for tunneling to become the dominant ionization mechanism can be estimated from the Keldysh γ parameter [17] $\gamma=\sqrt{I_p/2U_p}$, where I_p is the field-free ionization potential. A value of γ equal to unity roughly separates the intensity regions of multiphoton ($\gamma>1$) and tunneling ionization ($\gamma<1$) [18]. A direct comparison between the observed Ne spectrum and published data on the extent of the plateau (obtained with the same laser) [3] shows that the focused intensity in the Ne case was about 5×10^{14} W/cm^2. This intensity and the ionization potential of Na^+ and K^+ give γ values of 0.9 and 0.7, respectively. Hence we are just in the intensity region where tunneling is becoming an important mechanism for ionization and the I_p+3U_p rule should give a fair estimate of the extent of the plateaus.

The ionization potentials and the saturation intensities are higher for the alkali-metal ions than for Ne. Therefore, assuming that the I_p+3U_p rule is valid, one expects that for a given intensity, the plateau in these ions should extend at least as far as in Ne (even if the conversion efficiency for any harmonic in the plateau might be much lower). Our results, however, show that with a given laser-pulse energy and focusing, the harmonic spectrum obtained in the Ne gas jet extends to much higher orders. It thus appears as if the alkali-metal ions experienced a lower laser peak intensity than the Ne atoms.

D. Defocusing of the fundamental laser beam due to ionization

The peak intensity in the focus, in the absence of defocusing effects, was estimated from the extent of the Ne spectrum to be approximately 5×10^{14} W/cm^2. This is more than two orders of magnitude higher than the saturation intensity for the ionization of neutral Na [19]. Neutral atoms in the plasma are therefore ionized early during the rise time of the laser pulse. The ionization will take place first in the central part of the focal volume, close to the optical axis. As time progresses, the ionization will spread further out. This will lead to a radial gradient of free electrons, early during the rise time of the laser pulse. The refractive index of these free electrons results in a defocusing of the laser beam [20]. This defocusing prevents the laser beam in the plasma from reaching the same peak intensity as would have been reached in vacuum or in the Ne gas.

To study the role of ionization-induced defocusing under conditions similar to our experimental conditions, we modeled the propagation of the laser beam, using a computer code [21] taking space- and time-dependent ionization and free-electron dispersion into account. These results will be published elsewhere. The calculations were performed for atomic densities in the range of $10^{17}-10^{18}$ atoms/cm^3 and with a focused laser intensity (in the absence of defocusing) up to 50 times the saturation intensity for the neutral atoms. The results show significant defocusing effects and corresponding reductions in the peak intensity in the medium. The waist of the defocused beam is shifted in the direction of the focusing lens. One example of our numerical results is given as an illustration in Fig. 6. This particular result was obtained for a 6-mm-long medium and a 10-mm-long confocal parameter and the peak intensity, without defocusing, was 10 times I_{sat} for the neutral medium. The reason why we did not observe harmonic orders as high in the alkali-metal ions as in the neutral Ne gas might simply be due to this effect—the ions experienced much lower peak intensities. Since the free-electron dispersion is proportional to the square of the wavelength, ionization-induced defocusing can be expected to be considerably reduced if the laser is frequency doubled before focusing or if an excimer laser in the uv is used. Using a short-wavelength laser for the plasma production, e.g., a KrF-excimer laser, the plasma can be made essentially fully (singly) ionized directly during the plasma production stage [11]. Ionization-induced defocusing of the terawatt laser beam will therefore be reduced, leading to a higher focused intensity in the plasma.

E. Angular distributions

The spatial, or angular, distribution of the harmonic radiation is of great importance regarding the possibility of refocusing the radiation for use in various applications. The angular distribution of the harmonics observed in the present experiment was far from any ideal Gaussian. A detailed discussion of these structures is outside the scope of this paper. However, they are probably due to the free-electron dispersion in the focal region. This dispersion affects the propagation of the fundamental radiation, as discussed above, and hence the nonlinear polarization in the medium. The ionization-induced gradient in elec-

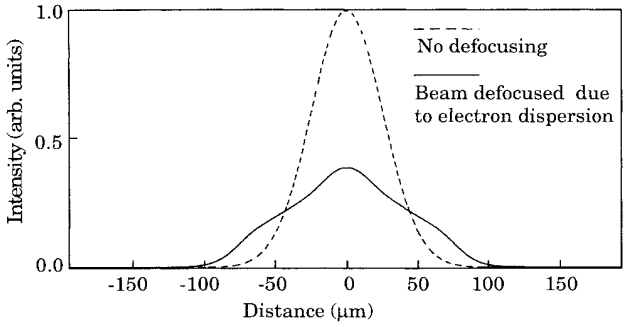

FIG. 6. Intensity at the focus as a function of transverse distance from the optical axis, illustrating ionization-induced defocusing of the fundamental laser beam in the alkali-metal plasma. The peak intensity for this modeling, in the absence of defocusing, was chosen to be 10 times the saturation intensity of the neutral atoms remaining in the plasma.

tron density also leads to radially dependent phase matching of the harmonics. This gives rise to radial variations in the phases of the harmonic fields at the end of the medium. These phase oscillations have been shown [22] to give rise to structures in the far-field spatial distribution of the harmonics, similar to those observed in this experiment.

V. SUMMARY AND CONCLUSIONS

We have studied high-order harmonic generation in laser-produced Na^+ and K^+ ions. The highest harmonic orders observed were not as high as expected from estimates based on the saturation intensities for these ions and on the focused intensities in the absence of defocusing. The spatial far-field distribution of the harmonic radiation was found to exhibit ring structures which varied with the focusing condition. The absolute value of the harmonic intensity is expected to be strongly dependent on phase matching and on the electron dispersion in particular. However, the rapid drop in intensity with harmonic order is interpreted as being due to the single-atom (-ion) response of the medium, i.e., corresponding to the cutoff of the characteristic plateau. The limited extents of the plateaus may, at least partly, be a consequence of ionization-induced defocusing of the high-power laser beam, reducing the peak intensity obtained in the medium. This defocusing is expected to be reduced by using a laser with a shorter wavelength for the harmonic generation or by using a laser in the uv for the plasma production, reducing the density of neutral atoms in the plasma.

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