RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 48, NUMBER 6 DECEMBER 1993

High-order harmonic emission from mixed fields

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(Received 7 September 1993)

Production of sum-frequency radiation and "even" harmonics from the mixed 1ω (1053 nm) and 2ω (527 nm) field is observed and compared to measurements of the relative conversion efficiency of the independent fields. By controlling the relative polarization of the two fields, we can control the mixing efficiency and produce coherent extreme ultraviolet (XUV) radiation polarized orthogonally to the strong driving field. Extension to produce XUV radiation of arbitrary polarization is discussed.

PACS number(s): 42.65.Ky, 32.80.Rm

Nonperturbative, high-order harmonic generation is a new technique for the production of coherent soft-x-ray radiation. Harmonic radiation extending to 7.8 nm has been observed in the interaction of intense infrared radiation with neon [1,2] and in this paper with helium. Production of shorter-wavelength radiation with a single visible or near-infrared laser field will require harmonic generation from ions [3,4]. Unfortunately, the efficiency of harmonic generation from ions will be severely limited by the deleterious effect of the plasma dispersion on phase matching. An alternative approach would be the use of a very-long-wavelength laser as suggested by the scaling of the harmonic cutoff as $V_{\mathrm{IP}}+3.2U_p$, where $U_p=9.33\times10^{-14}$ $[I(\mathrm{W/cm^2})][\lambda(\mu\mathrm{m})]^2$ and V_{IP} is the field-free ionization potential of the atom [3]. However, both quantum-mechanical [5], and classical [5,6] theories predict significantly decreased conversion efficiency in the plateau region with increasing wavelength. In this Rapid Communication, we present measurements of the relative conversion efficiency of 1053- and 527-nm fields independently and mixed together to produce high-order harmonic radiation. Mixing of a 1ω (1064 nm) with a strong 2ω (532 nm) field has been shown previously to have a significant effect on above-threshold ionization [7,8].

We find that the efficiency of the sum and difference frequency generation can be controlled by manipulating the relative polarization of the two fields. Sum frequency and "even" harmonics can be produced with an efficiency equal to odd harmonic generation of the more efficient, short-wavelength field. The use of two fields of arbitrary frequency and polarization gives us an ability to manipulate the production and polarization of harmonic radiation beyond that achievable with a single field.

The experiments utilize a Nd:phosphate glass-based laser system which can produce up to 10 J in a 600-fsec pulse at 1053 nm [9]. Type-I second harmonic generation in 98%-deuterated KDP produces up to 4.5 J in a 500-fsec pulse at 526 nm. In the experiments here, the power amplifiers have not been used, resulting in a maximum output of 1.5 J at 1053 nm and 300 mJ at 526 nm. This radiation is focused into a high pressure jet producing a helium density of 1×10^{19} atoms/cm^3 [10]. The jet is axially symmetric with an approximately Gaussian density distribution (diameter ≈750 μm) formed by a Mach 8 expansion into a vacuum. Harmonic and recombination radiation are dispersed by an imaging x-ray spectrometer which produces a flat-field spectrum on the photocathode of an x-ray streak camera. Laser light is focused into the jet by a 2-m focal-length aspheric singlet lens.

Harmonic spectra of the 1ω and 2ω fields independently at a peak irradiance of 4×10^{15} W/cm^2 are shown in Fig. 1. To produce this plot we have corrected the raw spectra for spectrometer and photocathode efficiencies. The signal-to-noise ratio of the raw data ranges from 10^5 for the lower-order harmonics to 2 for the 125th–129th orders. Under the conditions of this experiment, the harmonic spectrum is strongly dominated by generation from neutral species at intensities near the saturation intensity of 6.4×10^{14} W/cm^2. The "plateau" for the 526-nm field drops an order of magnitude from the 13th to 25th harmonic followed by a rapid decay to the 31st harmonic. This spectrum is in good agreement with single-atom calculations by Krause, Schafer, and Kulander [3]. Harmonics of the 1ω field are produced with approximately an order of magnitude reduced efficiency compared to those of the 2ω field until the 2ω cutoff. Beyond the 69th harmonic, the conversion efficiency remains virtually constant to the 129th. The extent of the plateau at 1053 nm is less than would be predicted by the $V_{\mathrm{IP}}+3U_p$ single-atom scaling at our saturation intensity of 9×10^{14} W/cm^2.

The 1ω and 2ω harmonic spectra were acquired under identical conditions of weak ionization and where the confocal parameter b is much greater than the length of the interaction region L. Integration over the emission volume (phase matching) predicts that under these conditions, the harmonic yield scales approximately as λ, i.e., favoring the longer wavelength. Hence, the difference in the emission strengths of the 1ω and 2ω harmonic fields in the plateau region (Fig. 1) suggests that the single-atom emission strength is a factor of 20 less at 1053 nm than at 526 nm until the 526-nm cutoff at 70 eV.

By combining the two fields, recent quantum-mechanical and classical calculations [5] suggest that it might be possible to increase the efficiency of harmonic production past the 2ω cutoff. Spectra obtained with the 2ω field only, the 1ω field orthogonal to the 2ω field, and the 1ω field parallel to the 2ω field are shown in Figs.

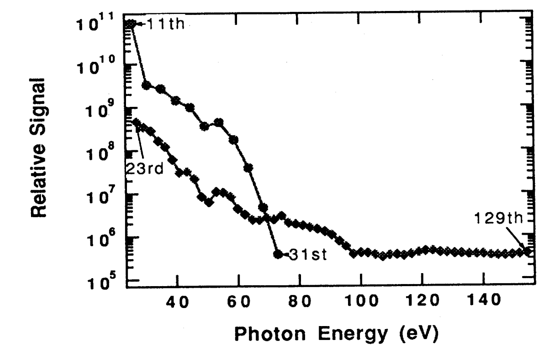

FIG. 1. Harmonic spectra produced in helium at $I > I_{sat}$ by a single 526-nm field (circles) and a single 1053-nm field (squares).

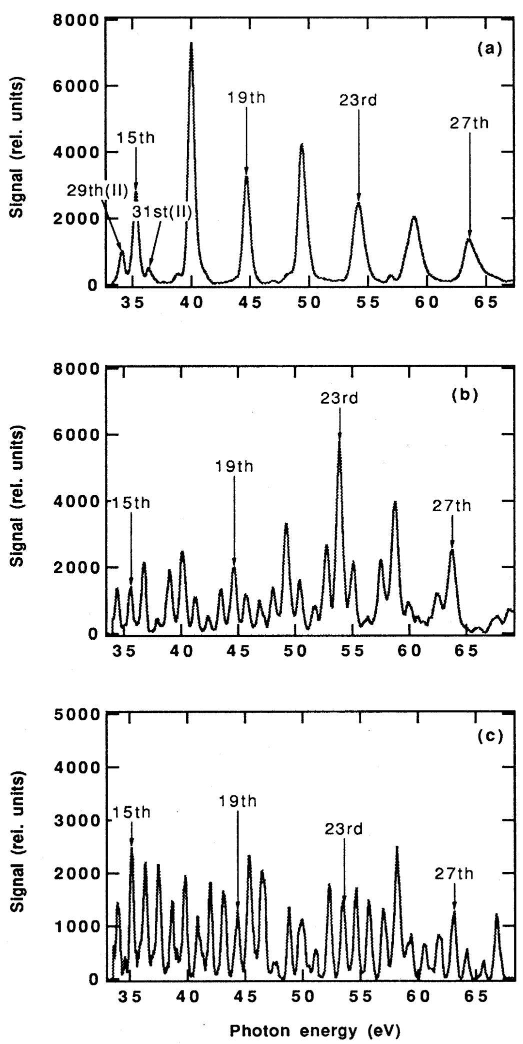

FIG. 2. Partial harmonic spectra obtained from (a) strong 526-nm (2ω) only, (b) strong 526-nm field and weak orthogonally polarized 1053-nm (1ω) field [$I(\omega) \leq 2\times10^{13}$ W/cm^2], and (c) strong 526-nm field and weak parallel polarized 1053-nm (1ω)field [$I(\omega) \leq 2\times10^{13}$ W/cm^2].

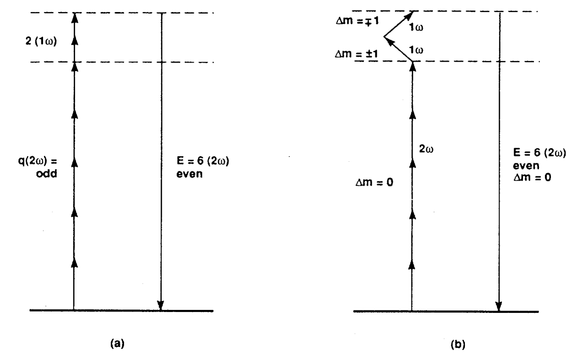

FIG. 3. Dipole-allowed pathway for the production of "even" harmonics of a strong 2ω field in the presence of a weak 1ω field: (a) parallel polarized fields, (b) orthogonally polarized.

2(a)–2(c), respectively. The secondary maxima on either side of the 15th harmonic peak of Fig. 2(a) are merely the 29th and 31st harmonics appearing in second order of the grating. These spectra were obtained with a strong 2ω field (1×10^{16} W/cm^2) and only a weak 1ω field (2×10^{13} W/cm^2). At this irradiance, no harmonic radiation is observed for the 1ω field by itself. This difference in intensity is due to chromatic dispersion in the lens producing group velocity walkoff and a focal-length difference of 5 cm between the 1053-nm pulse and the 526-nm pulse. The total group velocity walkoff between the two pulses as a result of all the optics is approximately 1.4 psec. We place the jet at the focus of the 526-nm pulse. Our previously measured saturation intensity for the ionization of helium at 526 nm (6.4×10^{14} W/cm^2) [11] occurs approximately 700 fsec before the peak of the 2ω pulse and 660 fsec after the peak of the 1ω pulse. The combination of the group velocity dispersion in the optics and the location of the jet 5 cm away from the focus for 1053-nm light results in an irradiance of 2×10^{13} W/cm^2 for the 1ω pulse at the time of maximum harmonic production by the 2ω field.

Between pairs of the usual odd harmonics of the 527-nm field are three additional peaks, which are observed only with the presence of the 1ω field [Figs. 2(b) and 2(c)]. The peak directly in between the odd harmonics of the 2ω field, which appears at an energy corresponding to an even harmonic, can be explained as true even-harmonic generation resulting from symmetry breaking of the Hamiltonian by the presence of the 1ω field. An alterna-

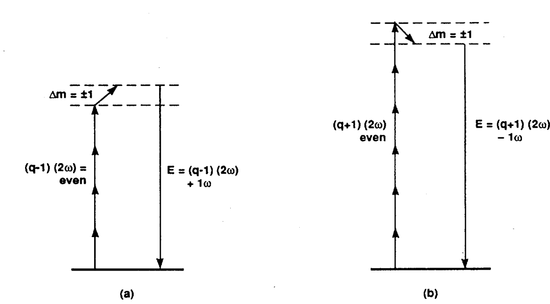

FIG. 4. Dipole-allowed pathway for the production of sum and difference harmonics of a strong 2ω field and an orthogonally polarized weak 1ω field.

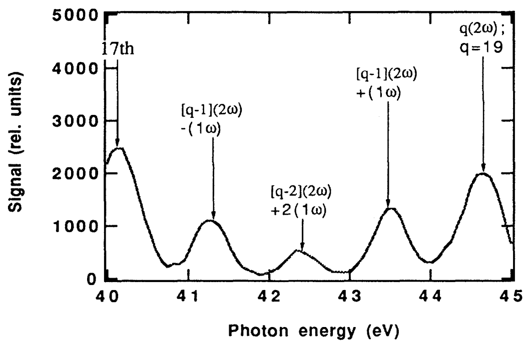

FIG. 5. Expanded spectra between the 17th and 19th harmonics of 2ω labeled with the lowest-order dipole-allowed pathway.

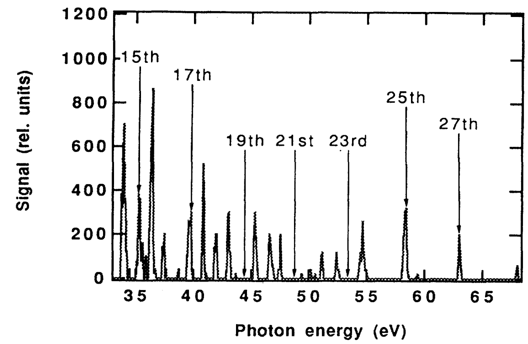

FIG. 6. A section of the mixed harmonic spectrum with an elliptically polarized strong (2ω)field. Labels indicate the location of odd 2ω harmonics.

tive explanation is that this peak results from the sum of an odd number of 2ω photons plus two 1ω photons. This sum-frequency process is dipole allowed ($\Delta m=0$) with parallel or orthogonally polarized fields (Fig. 3). In the case of orthogonally polarized fields, the dipole-allowed $\Delta m=0$ state is achieved by the first 1ω photon producing a $\Delta m=\pm1$ transition and the second photon producing a $\Delta m=\mp1$ transition relative to the $\Delta m=0$ state produced by the 2ω field. The overall state formed by the sum of an odd number of 2ω photons and an even number of 1ω photons is still $\Delta m=0$ relative to the ground state. With orthogonally polarized fields, this peak exhibits an efficiency approximately an order of magnitude below that of the usual odd harmonics of the 2ω field.

The secondary peaks on either side of the principal odd harmonics (1ω) of the 2ω field have an energy equal to odd multiples of the 1ω field. However, these are not 1ω harmonics in the usual sense in that they appear only when both fields are present. Figure 4 depicts the lowest-order dipole-allowed pathway that can produce this radiation. We attribute the peaks immediately on either side of the odd harmonics to an even number of 2ω photons plus or minus a single 1ω photon. The peak just below the normal $q\omega$ odd harmonic is formed by the absorption of $q-1$, 2ω photons of the strong 2ω field plus one additional 1ω photon. The peak just above the qth odd harmonic is formed by the absorption of $q+1$, 2ω photons minus one 1ω photon (Fig. 5).

This process is also dipole allowed ($\Delta m=\pm1$) with orthogonal fields, but is down by a factor of 3 in efficiency relative to the production of odd harmonics. These photons formed by the absorption of an even number of 2ω photons plus one orthogonally polarized 1ω photon should be polarized orthogonal to the strong 2ω field. Since the 1ω field is a coherent superposition of left and right circularly polarized photons, so too will be the mixed-field harmonics. When the fields are parallel polarized, the sum and difference frequency peaks including those involving the absorption of two 1ω photons are observed to have equal or even greater strength than the labeled odd harmonics [Fig. 2(c)] and are polarized parallel to the strong 2ω field.

The formation of mixed harmonics with orthogonally polarized light should enable the production of circularly polarized harmonic emission by having the weak field be circularly polarized. Unfortunately, with our equipment, it was not possible to produce pure circular 1ω light without inducing an unknown degree of ellipticity on the strong 2ω field. Ellipticity in the strong field changes the mixed-field spectrum dramatically (Fig. 6) with some harmonics eliminated completely, some increased.

In conclusion, we have measured the relative conversion efficiency of 1ω (1053 nm) and 2ω (526 nm) to high-order harmonic radiation in helium. The 2ω field is over an order of magnitude more efficient in producing harmonics in the plateau region but exhibits a plateau cutoff much earlier than the 1ω field, consistent with the $V_{\mathrm{IP}}+3U_p$ scaling predicted by Krause, Schafer, and Kulander [3]. By mixing a weak 1ω field with a strong 2ω field we can produce sum- and difference-frequency radiation including "even" harmonics. Although the strength of the 1ω field was too weak to cause the plateau to extend beyond the 2ω cutoff, the efficiency of the mixing products equaled the efficiency of the 2ω harmonics and was an order of magnitude stronger than the 1ω harmonics. The strength of the mixed-field emission exhibits a strong dependence on the relative polarization of the two fields. The use of orthogonally polarized fields produces harmonics with varying linear polarization depending on the order, while the use of circular polarization in the weak field should produce circularly polarized harmonic radiation.

We would like to acknowledge helpful discussions with P. Corkum, K. Kulander, H. Powell, and K. Schafer, and technical assistance by S. Herman and H. Nguyen. This work was performed for the U.S. Department of Energy under the auspices of Contract No. W-7405-Eng.

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