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Polarization-dependent high-order two-color mixing

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High-order frequency mixing experiments using the radiation of a high-power Ti:sapphire laser and its second harmonic are described and discussed. Linearly and circularly polarized light fields with comparable intensities have been used. For the theoretical description a three-dimensional quantum-mechanical calculation with a δ-function potential has been applied, showing quite good agreement with the experiments.

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A great deal of experimental and theoretical effort has recently been invested in the generation of high-order harmonics of a laser field incident on gaseous media. Besides being of interest *per se* the high harmonics yield access to a frequency range that is difficult to reach by other means. In the experiments high-power laser radiation with intensities well above 10^{14} W/cm^2, usually from Nd:glass [1], Ti:sapphire [2,3], or excimer [4] lasers, is focused preferentially into noble gases. In this way harmonics up to the 141st order and wavelengths as short as 7.5 nm have been achieved [5]. For the theoretical description of the harmonic generation process, different models and approaches have been developed [6], which in general allow for a stationary description of the experimental observations. For more detailed tests of theoretical models, high-order mixing experiments with two laser fields (two-color experiments) have gained some interest, due to additional degrees of freedom, such as relative polarizations, intensities, phases, and wavelengths. Furthermore, such experiments allow for the generation of broadly tunable xuv radiation as has been demonstrated recently [7].

In high-order harmonic generation experiments the power of the harmonics exhibits a characteristic "plateau" that ends with a fairly well-defined cutoff. The position of the cutoff can be understood, at least for a single atom, in terms of a simple classical model [8]. The model assumes that at some time the electron is released in the continuum, either by multiphoton or by tunneling ionization, with a velocity near zero. Subsequently, the electron is accelerated by the laser field. Depending on its time of release, the electron may return to the ionic core and recombine, emitting its energy in the form of one single harmonic photon. The maximum energy upon return determines the cutoff of the harmonic spectrum. This model works well for a monochromatic linearly polarized laser field. However, as soon as the polarization is no longer linear, the electron will, in general, never return to the core. Yet, high-order harmonic generation has been observed in such cases as well [9,10]. Studying high harmonic generation for nonmonochromatic fields with various polarizations will provide further clues to the physical mechanism of the cutoff. In addition, a better understanding might allow for some kind of "harmonic engineering" where the spectrum could be manipulated by judicious choice of the incident field. This provides part of the motivation of the present study.

High-order frequency mixing experiments reported so far [10,11] have only been performed with linear polarizations for both fields and with one field significantly weaker than the other. In this contribution we report on two-color mixing experiments with laser fields of comparable intensities and various relative polarizations, using the fundamental and frequency-doubled radiation of a short-pulse Ti:sapphire laser system. The experimental results are compared with numerical calculations using a δ-potential model [12].

The experimental setup is shown in Fig. 1. The pump laser is a 150-fs Ti:sapphire (BMI ALPHA 10A) operating at

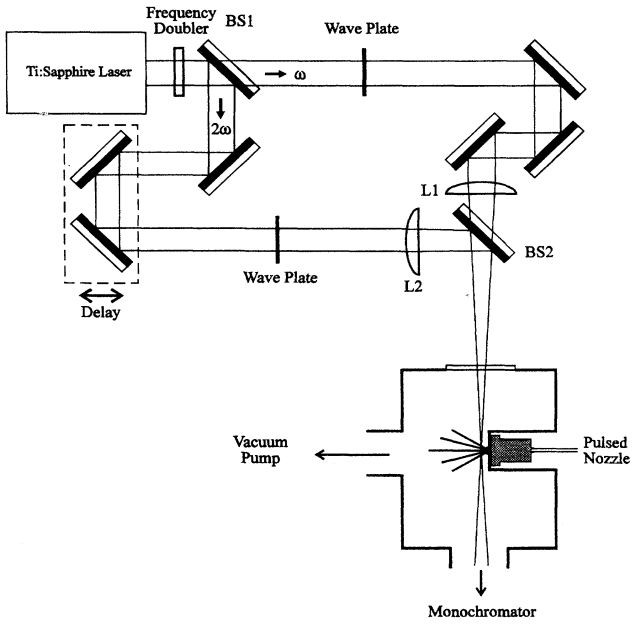

FIG. 1. Experimental setup. BS, beam splitter (highly reflective for 387 nm); L_1 and L_2, lenses with focal lengths of 40 and 50 cm, respectively.

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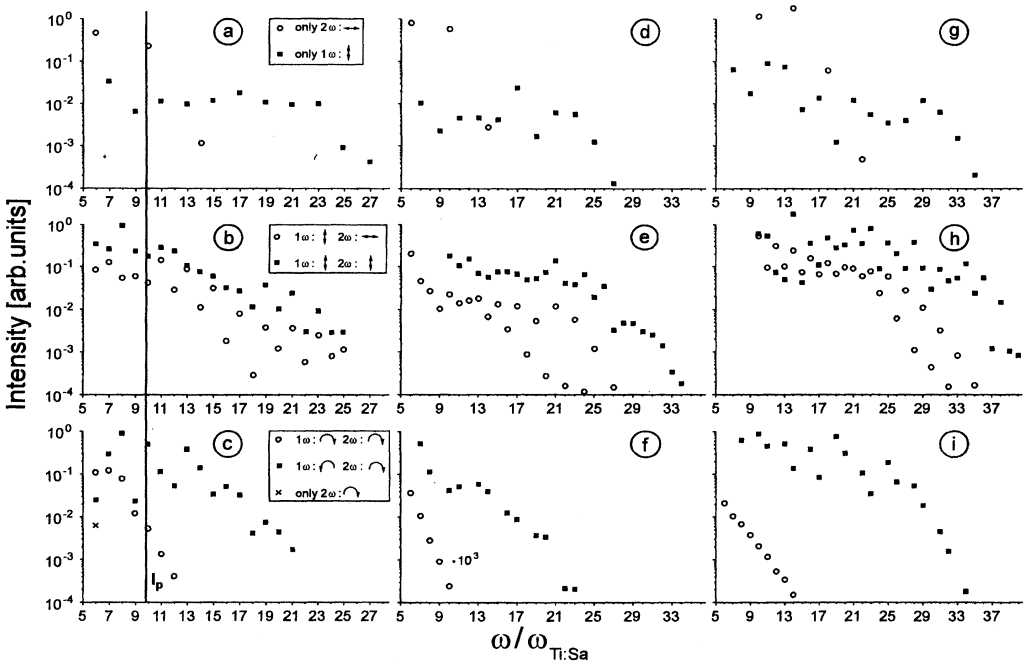

FIG. 2. $1\omega+2\omega$ frequency-mixing signals in argon for different relative polarizations of the fields in units of the photon energy of the pump laser (1.6 eV). The relative intensity scales are identical for (a)–(c) and for (d)–(i), respectively. (a)–(c) Experimental; I_P corresponds to the ionization potential of argon (15.76 eV). (d)–(i) Calculated intensities. (d)–(f) $I(\omega)=1.33\times10^{14}$ W/cm^2, $I(2\omega)=0.58\times10^{14}$ W/cm^2; (e) is averaged over the relative phase. (f) The results for the corotating circular polarizations have been multiplied by 10^3. (g)–(i) $I(\omega)=I(2\omega)=2\times10^{14}$ W/cm^2; (h) relative phase 180°. The experimental data points (a)–(c) are proportional to the photon numbers; the theoretical data points (d)–(i) give the quantity $|L|^2$ [12,16] which is proportional to the square of the dipolar moment.

773 nm. After frequency doubling, the remaining fundamental radiation is first separated by beam splitter BS1 and subsequently recombined by beam splitter BS2. With this approach (two focusing lenses and a variable delay between both pulses) a good temporal and spatial overlap in the focus could be obtained. The overlap was optimized by maximizing the intensity of a particular mixing signal ($2p2\omega$, see below) before each measurement. Furthermore, two waveplates could be inserted into the beams to manipulate the polarizations. Energies of about 12.5 mJ at 773 nm and about 6 mJ at 386.5 nm have been used. Due to the better focusability of the shorter wavelength beam, the intensities in the foci should be nearly the same. As nonlinear medium argon gas was injected into a vacuum chamber by a pulsed nozzle with a backing pressure of about 2 bar, corresponding to densities in the interaction region of 10^{17}–10^{18} cm^{-3}, the generated radiation was analyzed with a grazing-incidence monochromator (Jobin-Yvon LHT 30) and detected by a microchannel-plate detector.

To change the polarization of the beam, MgF_2 plates with the birefringent axis perpendicular to the surfaces have been used as zero-order waveplates. By a defined tilting of the plates the phase difference between the ordinary and extraordinary polarization could be changed continuously from zero to $\lambda/2$ and even further. The waveplates were aligned by means of standard film polarizers. With just one field on, we adjusted for circular polarization by reducing the intensity of the harmonics as much as possible [see Fig. 2(c)]. A remaining ellipticity is probably caused by the slight divergence of the beams.

With the described setup (Fig. 1) harmonic spectra for argon as nonlinear material have been recorded for the two pump fields 1ω and 2ω separately [Fig. 2(a)], for combined pump fields (1ω,2ω) with parallel or perpendicular linear polarizations [Fig. 2(b)] and with corotating or counterrotating circular polarizations [Fig. 2(c)]. In Figs. 2(d)–2(i) the experimental results are compared with calculations using a δ-function potential model as described below.

For the 1ω pump field alone [Fig. 2(a)] a clear plateau structure can be seen, extending from approximately the ionization energy I_P of argon (15.76 eV) up to a cutoff energy E_{cut} of about 40 eV. The measured cutoff energy agrees well with the equation $E_{cut}=I_P+\alpha U_P$ ($U_P\sim\lambda^2 I$, ponderomotive potential), using $\alpha=2.2$ [3,13] and assuming an intensity I given by the calculated saturation intensity $I_{sat}=2\times10^{14}$ W/cm^2 ($U_P=11$ eV for $\lambda=773$ nm), obtained from a field ionization model [14]. Experimentally, the intensities were about 3×10^{15} W/cm^2 and therefore well above I_{sat}. For the 2ω field alone, harmonics up to the seventh order ($7\times2\omega$) could be observed with no plateau structure, as the seventh harmonic of 2ω is already at the cutoff frequency. By rotating the polarization direction of the pump fields, the polarization sensitivity of the monochromator grating was tested, showing no substantial change of the signal intensity of the harmonics.

As can be seen from Fig. 2(a), the fundamental pump field ω generates odd harmonics at $(2p+1)\omega$, while the (2ω) field yields harmonics at $(2p+1)(2\omega)$ (p integer). Neither field generates the $2p2\omega$ even harmonics. With both pump fields acting simultaneously (with parallel or perpendicular polarization), "harmonics" at all higher multiples of ω are

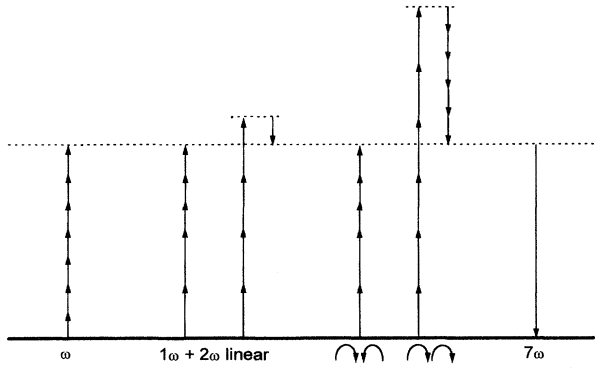

FIG. 3. Illustration of possible ways to generate radiation with a certain harmonic order (7ω) by mixing 1ω and 2ω fields with different polarizations.

possible, as indicated in Fig. 2(b). The $2p2\omega$ harmonics missing in Fig. 2(a) can now be generated by several mixing processes with $(2p-1)(2\omega)+2\omega$ as lowest-order process. In comparison with Fig. 2(a) the intensity of the odd harmonics of the 1ω field is significantly increased by up to two orders of magnitude, especially in the first half of the plateau. This can be explained by the fact that these harmonics can now be generated by different nonlinear processes, as indicated in Fig. 3, and that predominantly lower-order mixing processes may contribute.

Figure 2(b) shows that the harmonics generated by the parallel incident fields are consistently more intense than those generated by two perpendicular fields. This can be made plausible in several ways. First, this is what the above-mentioned classical model suggests: if the two fields do not have the same polarization, the electron virtually never returns to the position it started from. Hence, according to this model, there would be no harmonic emission at all were it not for wave packet spreading and electrons released in the continuum with nonzero initial velocities. While this argument depends on the legitimacy of the classical model, an analysis based on the combinatorics of perturbation theory (not necessarily of lowest order) leads to the same conclusion. One may show that

$$\frac{\chi^{(2n-1)}_{yy\cdots yyxx\cdots xx}}{\chi^{(2n-1)}_{xx\cdots xx}}=\frac{(2m)!(2(n-m))!n!}{m!(n-m)!(2n)!}, \qquad (1)$$

where the $\chi^{(2n-1)}$ in the numerator has $2m$ y components and $2(n-m)$ x components. The χ's operate on $2n-1$ fields $\mathbf{E}=E(\omega)\hat{\mathbf{x}}+E(2\omega)\hat{\mathbf{y}}$ or $\mathbf{E}=[E(\omega)+E(2\omega)]\hat{\mathbf{x}}$ for perpendicular and parallel polarizations, respectively. This causes the well-known degeneracy factors to occur, which, however, are identical for the two above cases and therefore do not affect the ratio if χ's are compared that correspond to the same harmonic frequency. Notice now that ratio (1) is smaller than 1 for all $m<n$, the more so the larger the n, and the harmonic intensities are proportional to this ratio squared. Furthermore, it can be shown that $\chi^{(n)}_{x\cdots xy\cdots y}=0$, if the number of x's or y's is odd. Therefore, the generated field has the same polarization as that pump field, which contributes an odd number of photons to the process, as confirmed by the nonperturbative theory described below [15].

Harmonics generated by two circularly polarized co- or counterrotating fields are shown in Fig. 2(c). With two circularly polarized fields the number of photons absorbed from or emitted to both fields must differ by 1 in order to fulfil angular momentum conservation. For oppositely polarized fields a given frequency can be generated to comparatively low order by the sum-mixing process $p(2\omega)+(p\pm1)\omega=(3p\pm1)\omega$. Therefore the frequencies $3p\omega$ are not allowed. In Fig. 2(c) the intensity of these frequencies is very low but not zero, which is probably caused by the imperfect circular polarization of the (2ω) field. An idea of the amount of deviation from pure circular polarization can be gotten from the cross ($\times$) mark in Fig. 2(c) that gives the intensity of the $3(2\omega)$ harmonic, which should be absent for pure circular polarization. In fact, its intensity is two orders of magnitude below the $3(2\omega)$ intensity for linear polarization [Fig. 2(a)], which corresponds to an ellipticity of about 0.9 (compare, e.g. [16]). Odd harmonics higher than this have not been detected. From angular momentum conservation it follows that the mixing field is circularly polarized with a polarization identical to that field which contributes the larger number of photons. On the other hand for equal polarizations higher-order difference-frequency mixing processes $(p\pm1)(2\omega)-(p\pm2)\omega=p\omega$ are required, as indicated in Fig. 3. Therefore, the signals for equal circular polarizations are less intense and more strongly decreasing with harmonic order, but all frequencies $p\omega$ can be generated. Now two paths (of different order) exist for each mixing frequency and therefore the polarization is different from either incident field.

For a more detailed theoretical description, the Schrödinger equation of the atom in the field of both lasers has to be solved and the propagation of the harmonics in the medium must be considered. Here we will restrict ourselves to the single-atom effects. Our major interest lies in the comparison of the various polarizations, and it appears likely that propagational effects would modify all of these cases in the same fashion. In our model, a three-dimensional δ-function potential has been used to describe the atomic binding potential [12,16]. The main advantage of this model is the fact that due to its three-dimensionality laser fields of arbitrary polarization can be considered about as easily as linearly polarized ones. The atom is characterized by just one parameter, which is related to the energy of the single bound state of the δ-function potential. It has been argued [16] that, for a given atom, it is more logical to adjust this energy to the energy difference between the ground state and the first excited state of the atom rather than to the ionization energy of the ground state itself. Hence, we use 11.6 eV in place of the actual binding energy of argon (15.6 eV). The calculation of the harmonic intensities proceeds analytically up to one final quadrature which has to be carried out numerically. The procedure is largely parallel to the case of one monochromatic linearly polarized laser field. Here we will just display the results. Details of the calculation will be given elsewhere [15].

We will show results of the model for two sets of parameters. First (case 1), we determined the intensities of the two laser fields such that they gave an optimal description of the one-color spectra. These values are $I(\omega)=1.33\times10^{14}$ W/cm^2 and $I(2\omega)=0.58\times10^{14}$ W/cm^2, which were then

used for the other cases. Second (case 2), we used the saturation intensities estimated from the field ionization model, viz., $I(\omega)=I(2\omega)=2\times10^{14}$ W/cm^2. The results are shown in Figs. 2(d)–2(f) (case 1) and Figs. 2(g)–2(i) (case 2). With the exception of the corotating circular polarizations, case 1 reproduces the cutoffs of all of the spectra very well. Since the δ-function potential always yields a one-color cutoff at approximately I_P+3U_P [16], case 2 then exhibits the one-color cutoff at harmonic orders higher than observed and this carries over to the two-color spectra. However, one may argue that in the relevant intensity regime the observed cutoff ought to be lower than the theoretical single-atom cutoff, owing to collective effects [3,13], so that case 2 can be compared to the data only for the lower parts of the plateau. If this is kept in mind, then both cases provide a fair agreement with the data. The notable exception is the case of corotating circular polarizations, where the calculated intensities are several orders of magnitude below the data. At least partially this can be explained by the imperfect circular polarization of the 2ω field. We should mention that the suppression of harmonics produced by the corotating circularly polarized fields versus the counterrotating fields is not a general feature; it does not occur when $U_P(\omega_1)/\omega_1 \sim U_P(\omega_2)/\omega_2$ and/or $\omega_2 \gg \omega_1$ [15].

The harmonic spectra produced by the two linearly polarized fields depends on the relative phase (as opposed to the two circular polarizations where they do not). The results given in Fig. 2(e) have been averaged over this phase while Fig. 2(h) is for a relative phase of 180° [electric fields proportional to $\sin(\omega t)$ and $-\sin(2\omega t)$].

In conclusion, most of the qualitative features of the experimental data agree with theoretical results, in particular. (i) Two parallel linearly polarized fields produce more intense signals than two perpendicularly polarized fields throughout the entire frequency regime. (ii) The harmonics produced by the perpendicularly polarized fields exhibit a characteristic even-odd behavior, the odd harmonics being more intense than the even ones. Notice that according to the theoretical model the odd harmonics for the perpendicular-linear-polarization case approach the one-color (1ω) harmonics in the upper part of the plateau. (iii) In a certain frequency region above I_P the mixing signals are stronger than the one-color harmonics by one or two orders of magnitude. (iv) As opposed to naive expectations derived from the above-mentioned classical picture, noncollinear polarization configurations produce mixing signals that are equally or even more intense than the harmonics of linearly polarized monochromatic fields.

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