Two-Color Phase Control in Tunneling Ionization and Harmonic Generation by a Strong Laser Field and Its Third Harmonic

S. Watanabe, K. Kondo, Y. Nabekawa, A. Sagisaka, and Y. Kobayashi
Institute for Solid State Physics, University of Tokyo, Roppongi 7-22-1, Minato-ku, Tokyo 106, Japan
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Two-color phase control in the total ion yield of ionization is successfully demonstrated in a tunneling regime by using a 100-fs Ti:sapphire laser and its third harmonic. Adding the third harmonic with an intensity of only 10% enhances the ion yield by a factor of 7. In photoelectron spectra, above threshold ionization peaks due to the third harmonic disappear when two colors are superimposed, resulting in a continuum spectrum. This shows two-color interference clearly. The intensities of high-order harmonics in the plateau region are enhanced by an order of magnitude.

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Multiphoton processes in a strong optical field, such as ionization and harmonic generation, have attracted both experimental and theoretical interest in recent years. Two-color phase control in ionization has been investigated both experimentally [1–3] and theoretically [4–6]. Recently two-color phase control was demonstrated in a relative height of above threshold ionization (ATI) peaks [1] and an asymmetric photoelectron angular distribution [2] by a fundamental laser beam (ω) and its second harmonic (2ω) or in two-path ionization by ω and the third harmonic (3ω) [3]. These experiments were performed in a multiphoton regime at a relatively weak field with long pulses (100 ps to 15 ns).

Recently, in the tunneling ionization regime, a very simple physical picture called a two-step model or a returning electron model [7,8] was proposed to explain such important phenomena as nonsequential double ionization and the cutoff of high-order harmonics. From this model, although the range of the validity is still being debated, not only the enhancements of high-order harmonics and of ion yield but also the extension of the cutoff order of harmonics are anticipated when two optical fields of different frequencies are coherently superimposed.

In this paper, the experiments of ionization and harmonic generation have been performed in a tunneling regime by ω and 3ω with 100-fs pulses. In this regime the effects of two-color interference are more drastic, and evident even in the absolute yields of ions and harmonics rather than in the relative change of ATI peaks or angular distribution. Furthermore, the effects are more exaggerated in the $\omega - 3\omega$ experiment than in the $\omega - 2\omega$ experiment. Even slight addition of 3ω to ω (10%–20% in intensity) enhanced the total yields of ions and harmonics by an order of magnitude. The periodic variation in the total ion yield was observed with a relative phase shift of 3ω to ω. The ATI peaks due to 3ω disappeared completely with the addition of ω, resulting in a continuum spectrum, thus showing the interference of two colors. These results are compared with the quasistatic model [9] and the two-step model.

The fundamental beam (745 nm) was generated by a 100-fs, sub-TW Ti:sapphire laser [10]. A single frequency-doubled YAG laser was used instead of a dye laser [10] to pump a three-stage amplifier chain at 10 Hz. Efficient frequency trippling was already reported in Ref. [10]. It should be noted that even at the exit of the mixing crystal 3ω is substantially delayed relative to ω in a 100-fs region. Taking into account the group delay in the window of the target chamber and the polarization direction, a delay line of ω and 3ω similar to the frequency trippler described in Ref. [10] was used. The polarization of 3ω was changed by a 90° rotator. The pulse envelopes of ω and 3ω were overlapped by adjusting the delay so as to generate the maximum 2ω signal in a thin mixing crystal after the window. The phase difference between ω and 3ω was changed by a piezoelectric transducer. The pulse widths of ω and 3ω were, respectively, 130 and 270 fs. The two coaxial beams of ω and 3ω were focused by an off-axis parabolic mirror ($f = 270$ or 125 cm) in a vacuum chamber to eliminate the color aberration and spherical aberration. The phase difference of two colors cannot be measured by a conventional cross correlator with a mixing crystal because the cross terms of ω and 3ω oscillate at an optical frequency (2ω) and give no stationary fringes. However, the phase difference was measured in the following experiments.

Before the experiments, the coherence of the ω beam was measured by a Michelson interferometer as shown in Fig. 1(a). The coherence of the output beam, which passed through many optics, was already degraded to a contrast (visibility) of 0.4. The total system was placed on a single table isolated from vibration with air cushions. Without this isolation, no fringes were observed. Because the contrast at the exit of an oscillator was 0.8, this degradation seems to be mainly due to the spatial phase distortion through many optics. The Michelson interferometer is, of course, used basically to measure the temporal coherence. But as the beam size increases up to 15–20 mm in diameter, the spatial phase distortion also degradates the contrast. Figure 1(a) gives

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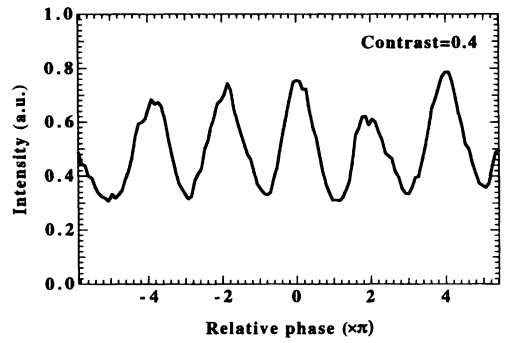
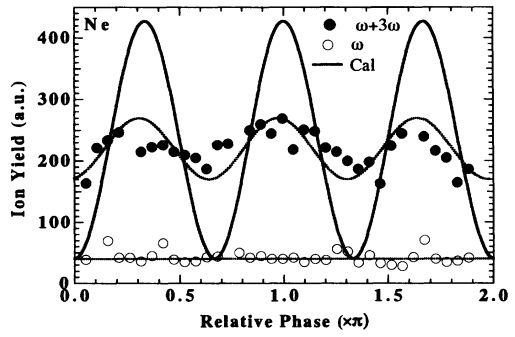


(a)



(b)

FIG. 1. (a) Michelson interferogram of the fundamental beam (745 nm) from the Ti:sapphire laser. (b) Ion yield of Ne^+ vs relative phase between ω and 3ω. The relative phase (ϕ) is defined by $E_\omega \cos(\omega t + \phi) + E_{3\omega} \cos(3\omega t)$. The intensity of ω and the field ratio ($E_{3\omega}/E_\omega$) are 5.7×10^{14} W/cm^2 and 0.4.

the calibration of the phase relative to the displacement of a piezoelectric transducer as well as the stability of the laser system. Figure 1(b) shows the ion yield of Ne^+ in relation to the phase difference between ω and 3ω. Ions were detected by a time of flight (TOF) analyzer [11]. One data point was averaged over 64 laser shots only when the 2ω energy, which passed through a turning mirror, was within 5% of the desired value. The ion yields due to two colors show a periodicity that is one-third of the Michelson fringes, and the average ion yield is enhanced by a factor of 6 over those of ω alone. The contrast of fringes is at best 0.3, which reflects mainly the initial contrast of ω. When 3ω was substantially delayed from ω, the ion yields were the same as those of ω. The ω intensity was set to 5.7×10^{14} W/cm^2, which is midway between the intensities of appearance and saturation. Note that the diffraction effects are different for the two colors, and thus the spot sizes and the confocal parameters are different. The phase difference is therefore not constant near the focus [1,3]. The volume effect can of course be considered in calculation, but it seems to lead to an extra complexity and rather to hide the essence. We therefore assumed that ionization occurs only near the most intense part of the laser focus [1]. The field of 3ω relative to ω was estimated as 0.4, which gives the average experimental ion yield. The solid curve shows the results of the calculation according to the quasistatic model [9]. As shown in Fig. 1(b) , the maximum ion yield should be an order of magnitude larger than that of ω at a relative phase of $\pi/3$, and the contrast of the fringes should be nearly equal to 1. In addition to the phase distortion of ω, the phase distortion in frequency conversion and focusing brings out further deviation from the ideal case. In the following experiments, we paid more attention to the average signal than to the phase dependence because the average enhancement due to the two-color interference is much more drastic than the contrast due to the phase variation.

In Fig. 2, the total ion yield of Ar^+ is shown as a function of field ratio ($E_{3\omega}/E_\omega$) at an ω intensity of 1.5×10^{14} W/cm^2. One data point is averaged over 128 shots selected by the 5% window of the 2ω energy. Above the field ratio of 0.3, the signals seem to saturate. This would be saturation of ionization where the time-integrated ionization probability becomes unity and no neutral atom remains. The phase-averaged calculation by the quasistatic model is fitted to the data with a field ratio shift of 1.2. The right-hand end of the solid curve shows the saturation. The average enhancement reaches 7 at a field ratio of 0.3.

Figure 3 shows the electron spectra in Ar at an ω intensity of about 3×10^{14} W/cm^2 and with $E_{3\omega}/E_\omega = 0.4$. The electron spectra were taken by a TOF method. Electrons were detected by a two-stage microchannel plate (MCP) 35 cm from the focal point and oriented in the direction of the polarization. The acceptance angle of the MCP was 3.6°. The flight path was shielded against the magnetic field of the Earth by a Mumetal tube. Data were analyzed by a digital signal analyzer (Tectronix DSA602), and were averaged over 128 shots selected by the 5% window of the 2ω energy. Figure 3 contains three curves due to ω, 3ω, and the coherent superimposition of

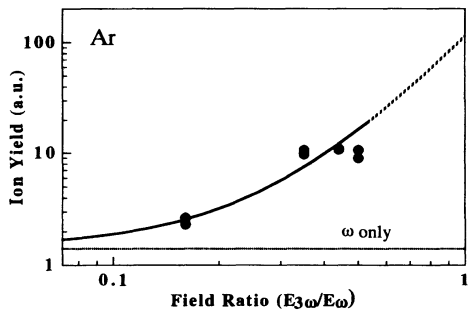


FIG. 2. Ion yield of Ar^+ vs field ratio. The solid line shows the result of calculation, and the dotted line shows the level of ion yield due to ω only. The intensity of ω is 1.5×10^{14} W/cm^2.

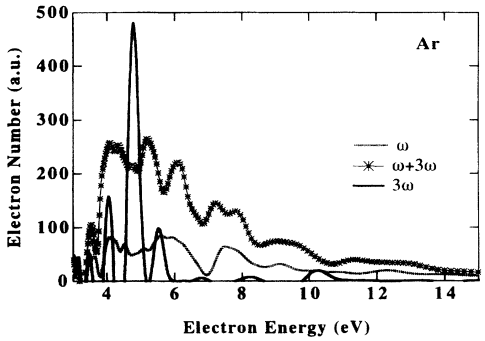


FIG. 3. Photoelectron spectra in Ar due to ω, 3ω, and two colors ($\omega + 3\omega$). The intensity of ω and the field ratio are 3×10^{14} W/cm^2 and 0.4.

ω and 3ω. The significant feature of this figure is the disappearance of the ATI peaks due to 3ω alone when two colors are superimposed. This is clear evidence of two-color interference. The spectrum due to two colors is wholly unlike the spectrum resulting from simple addition of the individual spectra due to ω and 3ω. Now the question is why the ATI peaks vanished so completely in spite of the phase uncertainty. The transition from the multiphoton to the tunneling regime is insensitive to the phase difference, and occurs if the fields of two colors are superimposed regardless of the phase difference.

The ω spectrum is well fitted to the quasistatic model, but the two-color spectrum deviates from the phase-averaged calculation at the high energy region. The reason for this deviation is not clear. One possible reason might be that only part of the angular distribution was monitored in this experiment.

Figure 4 shows the harmonic distributions of ω and of two colors (ω and 3ω) at an ω intensity of 5×10^{14} W/cm^2 with $E_{3\omega}/E_\omega = 0.3$. The experimental setup was essentially the same as that described in Ref. [12]. The harmonic intensities in the plateau region were

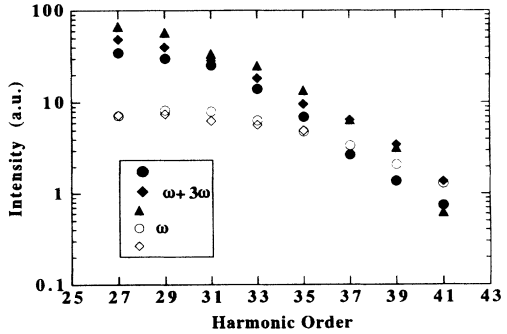


FIG. 4. Harmonic distributions in Ne due to ω (open symbols) and two colors (filled symbols). The intensity of ω and the field ratio are 5×10^{14} W/cm^2 and 0.3.

enhanced by an order of magnitude, whereas the explicit change of the cutoff order was not seen.

The distribution due to ω is typical. The dipole moment $|d(q\omega)|^2$ calculated by nonperturbative theory [13] and the two-step model predicts that the maximum energy of the harmonics is $I_p + kU_p$ ($k = 3.17$), where I_p is the ionization potential and U_p is the ponderomotive potential. In the experiment, the value of k tends to be between 2 and 3, depending on phase matching and propagation effects [14]. In this experiment, k was 1.8, and the relatively low cutoff order was probably due to tight focusing (the confocal parameter was 0.5 mm).

In two-color phase control, the two-step model predicts that at $E_{3\omega}/E_\omega = 0.3$ the value of k will vary between 2.8 and 3.7. This variation is out of the range that could be detected in this experiment, as already shown in Fig. 1(b), where the contrast observed was 0.3 rather than the predicted contrast of 1. This is why we observed no explicit change of the cutoff order. The phase-averaged calculation of $|d(q\omega)|^2$ shows the enhancement in intensity at the 27th harmonic, which is in the plateau region, to be by a factor of 7 (in good agreement with the experimental results).

The phase-averaged calculation of $|d(q\omega)|^2$ for a higher intensity at ω (7×10^{14} W/cm^2) with a higher field ratio (0.5) predicts the harmonic intensity to be enhanced by more than an order of magnitude and also predicts the cutoff order to be extended from the 83rd to the 103rd harmonic in Ne. Unfortunately this condition was not attained in the present experiment because of the low throughput of the delay line. The throughput of 3ω was only 30%. Improvement of the delay line will allow us to further increase harmonic intensity and cutoff order.

In conclusion, two-color phase control in the total ion yield of ionization has been demonstrated. At the same time, this experiment shows the degradation of the contrast (compared with the theoretical contrast) because of the initial phase distortion of ω along with the phase change between ω and 3ω during focusing. The two-color interference nonetheless enhanced the yields of ions and harmonics dramatically by an order of magnitude. The evidence of two-color interference was clear in the photoelectron spectra. The phase-averaged calculation according to the two-step model is compared with the observed yields of ions and harmonics in the framework of this experiment. But further work would be necessary to examine the range of the validity of this model. The next step will be to observe the extension of the harmonic order in an experiment with a higher field ratio. The results of the $\omega - 2\omega$ experiment will be compared with the present experiment [15].

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