Destructive Interference during High Harmonic Generation in Mixed Gases

Tsuneto Kanai,* Eiji J. Takahashi, Yasuo Nabekawa, and Katsumi Midorikawa
Laser Technology Laboratory, RIKEN, 2-1 Hirosawa, Wako-shi, Saitama 351-0198, Japan
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We report on the first experimental evidence of the destructive and constructive interference of high harmonics generated in a mixed gas of He and Ne, which facilitates the coherent control of high harmonic generation. Theoretically, we develop an analytical model of high harmonic generation in mixed gases and succeed in reproducing the experimental results and deriving the optimization conditions for the process. The observed interference modulation is attributed to the difference between the phases of the intrinsically chirped harmonic pulses from He and Ne, which leads to a novel method for broadband measurement of the harmonic phases and for observing the underlying attosecond electron dynamics.

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The high harmonic generation (HHG) in pure atoms using ultrashort lasers has been a subject of intense studies for its applications as a coherent attosecond radiation source in the extreme ultraviolet (XUV) and soft x-ray regions [1–4]. The physics of HHG in atoms, including the amplitude and the phase of harmonics, are well understood by the three-step model (Fig. 1) [5]: First, a part of the bound-state electron wave function tunnels through the potential barrier modified by the intense laser field and appears in the continuum (step 1). The freed electron wave packet is then driven by the laser field and after the field reverses its direction, it has a probability of returning to the parent ion with a quantum mechanical phase according to the excursion time τ_j of the trajectory j and the ionization potential I_p^i of the atoms i (step 2). The harmonic photon is emitted by the coherent oscillation between returning electron wave packet and the bound-state electron wave function at time t (step 3). The harmonics generated at the sites where the event happened by chance, interfering with each other, propagate through the gas medium under the influence of dispersion and absorption of the medium.

Characterizing high harmonic (HH) pulses and enhancing their efficiency have been the most important issues in ultrafast optical science not only for investigating its generation process but also for using it as a scientific tool to explore attosecond physics. The loosely focused beam geometry under phase-matching conditions is one of the most successful method for enhancing the efficiency of HHG [6,7]. Based on the two-photon above-threshold ionization (ATI) [8], such intense harmonics provide us robust methods to measure the amplitude and the phase of the harmonics such as FROG (frequency-resolved optical gating) [9] and PANTHER (the photoelectron analysis with nonresonant two-photon-ionization for harmonic electric-field reconstruction) [4]. Harmonics that are not intense enough to induce nonlinear processes, on the other hand, were characterized by simultaneous use of intense infrared laser pulses. Hentschel *et al.* developed physics on electron dynamics in the combined field and demonstrated measurement of an isolated attosecond pulse [1]. Paul *et al.* focused on the quantum interference that appears in the two-color ATI and developed a method to measure relative phases of harmonics called RABITT (reconstruction of attosecond beating by interference of two-photon transitions [10]). Detailed understanding of the underlying physics will sometimes offer us new measurement and control methodologies.

Recently, it has been rediscovered that the three-step model has more physical structures than expected. Itatani *et al.* (with HHG [11]) and Kanai *et al.* (with HHG and ionization [12]) proposed novel methods to probe molecular structures with 100-attosecond temporal resolution using underlying physics of HHG. In 2006, Baker *et al.* applied the above technique to H_2, D_2, CH_4, and CD_4 molecules by assuming the theoretically calculated excursion times of the short trajectories τ_1 [13]. Measuring the excursion times in the attosecond region should be the next challenge in attosecond physics. For the excursion time in the femtosecond region, Salières *et al.* applied the so-called intensity dependent atomic dipole phase and by estimating laser intensity $I_{IR} = 1.35 \times 10^{14}$ W/cm^2, determined the excursion time τ_1 for the 19th harmonic to be 1.2 fs [14]. However, the uncertainty of laser intensities, which are difficult to control and determine with accuracy,

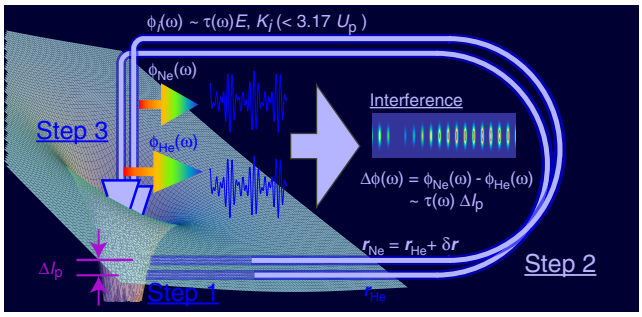

FIG. 1 (color online). An illustration of two quantum paths of electrons that contribute the HHG in a mixed gas. The variation on quantum path $\delta \boldsymbol{r}$ generates the phase difference of harmonics. Notice that ΔI_p expresses the difference in energy between the two potentials.

prevents researchers from precise measurement of τ_j using the intensity dependence [15].

In this Letter, by developing the three-step model for HHG in mixed gases, we measure the excursion times in the attosecond region in a novel and robust way. Experimentally, we obtain the first evidence of the DI and the constructive interference (CI) in a mixed gas of He and Ne, which facilitates the coherent control of HHG [16]. Theoretically, we develop an analytical theory of HHG in mixed gases by combining the propagation equation [6] and the saddle point method [5] and succeed in reproducing the experimental results and deriving the optimization conditions for the process. The observed interference modulation is attributed to the difference between the phases of the intrinsically chirped harmonic pulses from He and Ne, which offers a novel method for broadband measurement of the harmonic phases and for observing the underlying attosecond electron dynamics with no knowledge of laser intensity. Furthermore, our method can serve as a gaseous tunable bandpass filter in the XUV and soft x-ray regions, which keeps playing crucial roles in attosecond physics.

Consider the power spectra of the harmonics from gas 1, 2, and mixture of them, $\mathcal{P}_1(\omega)$, $\mathcal{P}_2(\omega)$, and $\mathcal{P}_{1+2}(\omega)$. As can be seen in the simplest case of two point emitters at the site 1 and 2, $|d_1(\omega)+d_2(\omega)|^2 = |d_1(\omega)|^2 + |d_2(\omega)|^2 + 2\mathrm{Re}[d_1(\omega)d_2^*(\omega)]$, where $d_i(\omega)$ is the harmonic amplitude at the site i, the effects of the interference between the two HHG processes (i.e., the counterpart of $2\mathrm{Re}[d_1(\omega)d_2^*(\omega)]$) should appear in the difference between $\mathcal{P}_{1+2}(\omega)$ and $\mathcal{P}_1(\omega)+\mathcal{P}_2(\omega)$. By integrating the propagation equation of harmonics and the Schrödinger equation of the active electrons, the spectrum $\mathcal{P}_{\Sigma i}(\omega)$ from the mixed gas composed of gases i's at the q-th harmonics ($\omega = q\omega_0 \equiv \omega_q$ with the photon energy of the fundamental field ω_0) emitted on axis can be derived as

$$\begin{aligned}\mathcal{P}_{\Sigma i}(\omega_q) &\propto (\rho_{\mathrm{tot}}L)^2|\langle d(\omega_q)\rangle|^2 \times [(2L_a(q))^{-2} + \pi^2 L_c^{-2}(q)]^{-1} \\ &\quad \cdot [1 + e^{-L_a^{-1}(q)} - 2\cos(\pi L_c^{-1}(q)) e^{-(2L_a(q))^{-1}}] \\ &\equiv \mathcal{I}_{\mathrm{gen}}(\omega_q) \times \mathcal{I}_{\mathrm{prop}}(\omega_q), \end{aligned} \tag{1}$$

where $\rho_{\mathrm{tot}} := \sum_i \rho_i$ with ρ_i being the density of the gas i, L is the medium length, $\langle\cdots\rangle$ denotes the density weighted average, e.g., $\langle d\rangle = \sum_i r_i d_i$ with $r_i := \rho_i/\rho_{\mathrm{tot}}$. $L_a(q) := [2\rho_{\mathrm{tot}} L \lambda_0 r_e \langle f_2\rangle/q]^{-1}$ is the dimensionless absorption length, where $\lambda_0 := 2\pi c/\omega_0$, r_e is the classical electron radius, and f_2 is the imaginary part of the atomic scattering factor. $L_c(q) :== \pi/(\Delta k_{\mathrm{tot}}(\omega_q)L)$ is the dimensionless coherence length with $\Delta k_{\mathrm{tot}} := k_q - qk_0$, where k_q and k_0 are the wave vectors of the q-th harmonics and of the fundamental field, respectively [17].

The first term in Eq. (1), $\mathcal{I}_{\mathrm{gen}}(\omega_q)$, expresses the generation of harmonics by integrated single atom responses interfering with each other. In the two-gas case, it can be simplified as $\mathcal{I}_{\mathrm{gen}}(\omega_q) = \rho_1^2|d_1(\omega_q)|^2|1 + r_2/(1-r_2)|d_2(\omega_q)/d_1(\omega_q)|e^{i\Delta\varphi(\omega_q)}|^2$, where $\Delta\varphi(\omega_q) := \varphi_2(\omega_q) - \varphi_1(\omega_q)$ is the relative phase of the harmonics from the two gases 1 and 2. Straightforwardly, the condition for the CI ($\mu = 0$) and the DI ($\mu = 1$) can be derived as $\Delta\varphi(\omega_q) = (2n + \delta_\mu^1)\pi (n \in \mathbb{Z})$, with the Kronecker delta δ_ν^μ. These interferences are the most fundamental phenomena of HHG in mixed gases; they always appear where the "two" spectra overlap. It should be noted that when the condition for DI is satisfied for the q-th harmonics, we can completely suppress HHG by merely setting the density ratio $r_2 = [1 + (d_2(\omega_q)/d_1(\omega_q))]^{-1}$ regardless of the propagation condition.

Here we restrict our attention to the simplest and ordinary case in which $d_i(\omega_q)$ can be well described by the nonlinear response to the fundamental pulse only. As can be confirmed later, this is in the case of a He-Ne mixed gas. If the density of generated harmonic photons in the interaction region is not low enough compared to that of the fundamental photons, $d_i(\omega_q)$ should strongly be influenced by the generated harmonic fields [18]. Using the saddle point method $\delta S_i(\boldsymbol{p}, t, \tau) = 0$, where $S_i(\boldsymbol{p}, t, \tau)$ is the action of the active electron in the atom i, the stationary phase S_i^s is found to be a linear function of I_p^i and the ponderomotive potential U_p: $S_i^s = \tau^s I_p^i + [\tau^s - 2(1-\cos\tau^s)/\tau^s - C(\tau^s)\cos(2t^s - \tau^s)]U_p$, where $C(\tau) := \sin(\tau) - 4\sin^2(\tau/2)/\tau$ [5,19]. Considering $U_p \propto I_{\mathrm{IR}}$, atomic dipole phases linearly depend on the laser intensity I_{IR} and the dependence attracted much attention for the potential to measure the harmonic phases [9,15,20,21] although the underlying parameter of laser intensity cannot be controlled precisely. We offer an alternative approach by using the other dependence, i.e., the dependence on I_p, which is the intrinsic quantity determined by nature exactly. By preparing two gases with different I_p's, we can measure the phase of harmonics in a robust way. In fact, when the short trajectory is selected as in our case [7], the relative phase $\Delta\varphi(\omega_q)$ takes a simple form [22,23],

$$\Delta\varphi(\omega_q) \sim \tau_1(\omega_q)\Delta I_p. \tag{2}$$

The first order term in ΔI_p comes from the difference of the potential energy, and the higher order terms should be included when $\Delta I_p/U_p$ is not small enough. The phases, which the freed electrons accumulate in their different quantum paths, are transferred to the harmonics through the coherent process of HHG and lead to the interferences (Fig. 1).

The dimensionless second term in Eq. (1), $\mathcal{I}_{\mathrm{prop}}(\omega_q)$, expresses the role of propagation in mixed gases. As in the case of HHG in pure gases [24], the optimization conditions, $1/3 > L_a$ and $L_c > 5L_a$ ensure that the macroscopic response is more than half of the maximum response. The asymptotic value in $L \to \infty$ and $L_c \to \infty$ is independent of ρ_{tot} and is a monotonically increasing function of $|\langle d(\omega_q)\rangle/\langle\sigma(\omega_q)\rangle|^2$, where $\langle\sigma(\omega_q)\rangle \propto L_a^{-1}$ is the expectation value of the ionization cross section. Optimizing HHG requires us to simultaneously maximize $|\langle d(\omega_q)\rangle|/\langle\sigma(\omega_q)\rangle$

and to fulfill the above conditions. Considering the interference effect on $|\langle d(\omega_q)\rangle|$, the optimization conditions for HHG in mixed gases are now given by $\Delta\varphi(\omega_q) \sim 2n\pi (n \in \mathbb{Z})$, in addition to the usual conditions $1/3 > L_a$ and $L_c > 5L_a$.

In order to test this model, we performed an experiment with a Ti:sapphire laser having the center wavelength $\lambda_0 \sim 800$ nm, the maximum energy $\sim$200 mJ, and the pulse duration $\sim$30 fs. The fundamental pulse was loosely focused with an $R = 10$ m concave mirror into an interaction gas cell filled with a pure and/or a mixed gas, and phase-matched harmonics were generated in the cell by balancing the Gouy phase and the negative dispersion of the gas medium. The generated harmonics were spectrally resolved with a flat-field grating and detected by a microchannel plate and a CCD camera. See Ref. [7] for the details of our setup. The intensity of the fundamental pulses was estimated to be $I_{\mathrm{IR}} \sim 4 \times 10^{14}$ W cm^{-1}, which is below the saturation of ionization of atoms, ensuring that HHs are generated mostly at the peak of the laser pulse. The medium length was set to be $L \sim 13$ cm.

Figure 2(a) shows the typical harmonic spectra in He ($p_{\mathrm{He}} = 13.5$ Torr), Ne ($p_{\mathrm{Ne}} = 1.5$ Torr), and the mixture of them ($r_{\mathrm{Ne}} = 10\%$, $p_{\mathrm{tot}} = 15.0$ Torr). The second-order diffraction rays of the 45th–71st harmonics appeared at the side of the 23rd–35th harmonics. As expected, an interference modulation is observed in the spectra from the mixed gas: The harmonics around 51st are enhanced, and those around 29th are suppressed. It should be emphasized that $\mathcal{P}_{\mathrm{He+Ne}}(\omega_{29})$ is completely suppressed compared to $\mathcal{P}_{\mathrm{He}}(\omega_{29}) + \mathcal{P}_{\mathrm{Ne}}(\omega_{29})$. Physically, this spectral modulation should correspond to the interference between the harmonics from He and Ne. In fact, with the help of the Eq. (1), the experimental results can be translated to the interference modulation characterized by relative phase $\Delta\varphi(\omega_q)$ of harmonics. Figure 2(b) shows the relative phase $\Delta\varphi(\omega_q)$ as a function of harmonic order q. DI and CI take place when $\Delta\varphi(\omega_q) \sim \pi$ and 0, respectively, and within the experimental errors due to the laser fluctuation and the ground state depletion, we see satisfactory agreement between the experimental results and the theoretical calculations, which ensures the validity of our interpretation and model.

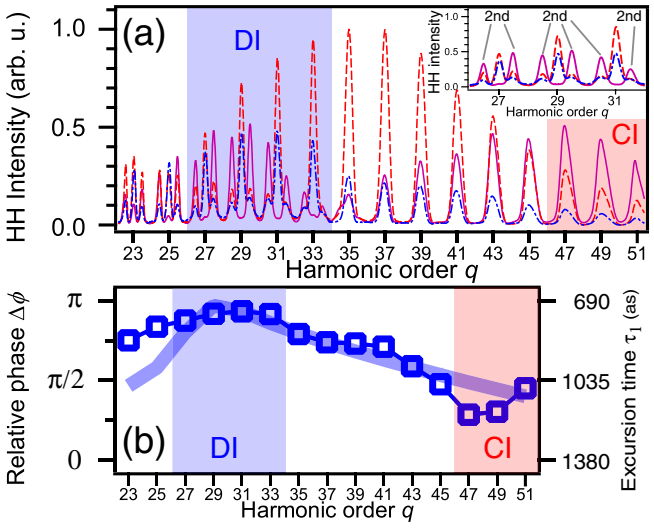

FIG. 2 (color online). (a) The harmonic spectra from a 13.5 Torr He gas (dashed line), a 1.5 Torr Ne gas (dashed-dotted line), and the mixture of them (solid line). Inset shows the enlarged illustration of the spectra, where DI takes place. (b) The experimentally determined relative phases of harmonics and the corresponding excursion time τ_1 as a function of harmonic order (squares). The error bars are about the sizes of the symbols for the experimental data. Theoretically obtained curve are also shown by the solid line.

One of the important consequences of these results can be revealed when one regards the observed interference as an inverse problem. From Eq. (2), observing the relative phase corresponds to the broadband measurement of the excursion time τ_1 and thus the estimation of the individual harmonic phase $\varphi_i(\omega_q)$ [14]. Measuring attosecond excursion times offers a crucial basis for the methods of probing the ultrafast dynamics of molecules by using electron wave packet [11–13,25]. Directly from the DI condition, we obtain $\tau_1(\omega_{29}) \sim 690$ as, which is the shortest duration of particle motion observed to date [2,14]. It should be noted that the attosecond dynamics of electrons in HHG can be realized only in high intensity laser field and at lower order harmonics. Accordingly, the excursion time of $\tau_1(\omega_{19}) = 1.2$ fs by Salières *et al.* using the lower intensity of $I_{\mathrm{IR}} = 1.35 \times 10^{14}$ W/cm^2 [14], is much longer than our results. Our approach, keeping the advantages of using interference phenomena which can be caused even by weak harmonics with no assumption of atomic physics in the measurement stage, minimizes the effect of path fluctuations. Because of the extremely short wavelength of harmonics, path fluctuations inevitably give effects even on the fine experiment by Corci *et al.* [15]. In our case, the "two" pulses, whose relative phase is of importance, propagate through the same path and phase fluctuations are then automatically cancelled [26]. The chirped relative phase required for spectral interference, which is easily controlled with a delay state and a nonlinear optics in the IR regions [27], is realized in a robust manner using the HHG process directly.

Another important consequence is the demonstration of tunable bandpass filtering using mixed gases. Bandpass optics have been crucial experimental elements in attosecond physics: They are used to compress harmonic pulses [10,20], to use harmonics as a tool for energy-dependent processes in matters [3], and to generate single attosecond pulses [1,2]. Usual bandpass optics, however, inevitably reduce the number of the desired harmonic photons during propagating through the dissipating material, and their band cannot be controlled easily. From Eq. (2), the energy where CI takes place is given by $\omega = \tau_1^{-1}(2n\pi/\Delta I_p)$, which directly indicates the dependence on ΔI_p in addition to the obvious dependence on I_{IR}. By

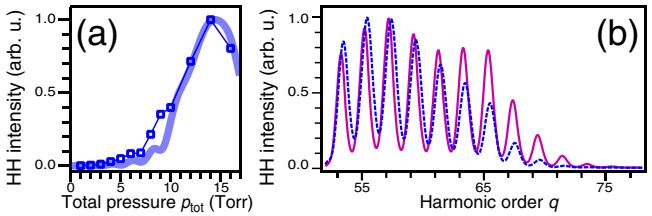

FIG. 3 (color online). (a) Integrated intensity of 59th harmonics from a He-Ne gas ($r_{Ne} = 10\%$) as a function of total pressure p_{tot}. (b) Harmonic spectra from the optimized He-Ne gas ($r_{Ne} = 35\%$ and $p_{tot} = 14.0$ Torr; solid line) and the optimized pure Ne gas ($p_{Ne} = 9.1$ Torr; dashed line).

using a pair of atoms or molecules with different ΔI_p, our method can be used for gaseous tunable bandpass filtering in the XUV and soft x-ray regions without passing any dissipating optics. While multilayer optics use interferences due to the path differences between the boundaries based on the fine deposition technologies, our novel filtering uses interferences due to the phase difference directly made from the underlying ultrafast electron dynamics in HHG.

To enhance HHG, on the other hand, both I_{gen} and I_{prop} should be optimized simultaneously. Figure 3(a) shows the integrated intensity of the 59th harmonics from a He-Ne gas ($r_{Ne} = 10\%$) as a function of the total pressure p_{tot}. The pressure that maximizes the harmonic intensity corresponds to the phase-matching pressure and the shape of the experimental curves is reproduced by our model, which indicate the validity of our model again and ensures the ansatz that $d_i(\omega_q)$ is dominated by the response to the fundamental pulse. Figure 3(b) shows the harmonic spectra from the optimized He-Ne gas ($r_{Ne} = 35\%$ and $p_{tot} = 14.0$ Torr) and the optimized pure Ne gas ($p_{Ne} = 9.1$ Torr) [7]. The optimized pressure $p_{tot} = 14.0$ Torr was determined by the phase-matching mechanism demonstrated in Fig. 3(a), the ratio $r_{Ne} = 35\%$ was determined experimentally, and the medium length $L \sim 13$ cm was set to satisfy the absorption limited condition [24]. For I_{gen}, the condition for CI is found to be relatively satisfied ($\Delta\varphi \sim 1$ rad) for harmonics with $q = 55$–65, and accordingly, the maximum pulse energy was achieved in this region; by comparing the harmonic intensity from the He-Ne gas to that from the Ne gas, the pulse energy of each harmonic ($q = 55$–65) was estimated to be (20–30) nJ/pulse, which are the maximum energy achieved to date in this region [7]. Furthermore, the extension of the cutoff was observed. This is another advantage of using mixed gases; the density of Ne, which are easily ionized compared to He, can be set to be lower than that of pure Ne, and as indicated by the blueshift of the spectrum $\mathcal{P}_{He+Ne}(\omega_q)$, the effective laser intensity can be enhanced due to the suppressed ionization.

As the third media with the possibility to supplant pure atoms and molecules, mixed gases can serve as a new route not only to control and characterize harmonics but also to measure the electron excursion times. Nearly half a century after the invention of the first continuous gas laser, the He-Ne laser [28], this fundamental mixture system, which is composed of the lightest two noble gases with appropriate energy structures, provides us a novel coherent radiation source again.

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*Electronic address: tkanai@riken.jp

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