Ionization-Induced Multiwave Mixing: Terahertz Generation with Two-Color Laser Pulses of Various Frequency Ratios

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Ultrafast strong-field ionization is shown to be accompanied by atypical multiwave mixing with the number of mixed waves defined by the dependence of the ionization rate on the field strength. For two-color laser pulses of various frequency ratios, this results in the excitation of a free-electron current at laser combination frequencies and possibly in the excitation of the zero-frequency (residual) current responsible for terahertz (THz) generation in a formed plasma. The high-order nature of ionization-induced wave mixing may cause THz generation with uncommon laser frequency ratios (such as 2:3 and 3:4) to be virtually as effective as that with the commonly used frequency ratio of 1:2.

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The ionization-induced generation of broadband intense terahertz (THz) radiation by ultrashort (femtosecond) laser pulses attracts considerable interest due to various applications. They include THz time-domain spectroscopy [1], the probing and control of various ultrafast processes and THz imaging with subwavelength resolution [2,3], and the study and use of nonlinear THz interactions in novel media such as graphene and semiconductor structures [4,5]. Most of the experimental and theoretical studies of such laser-plasma generation are now concentrated on the so-called two-color method providing strong THz pulses with a very broad spectrum [6–12]. This method usually employs two-color ionizing pulses that originate from a one-color ultrashort laser pulse (usually from a Ti:sapphire generator with a wavelength near 800 nm) passing through a frequency-doubling crystal [6–9]. Such pulses contain strong quasimonochromatic components at the main frequency along with weaker components at doubled frequency. The optical parametric amplifiers and their growing availability extend the range of two-color pulses to employ. So the recent works [10,11] considered THz generation in air ionized by a two-color pulse containing a strong main field along with a weak additional field tunable near the half value of the main frequency. And in Ref. [12], the available frequency tuning allowed determining the wavelength scaling of the THz yield and obtaining strong THz fields.

The detailed analysis in Ref. [9] proved that the main contribution to low-frequency THz radiation is defined by the free-electron response in the formed laser plasma. In this Letter, we examine for the first time how the intrinsic nonlinear features of this response lead to THz generation. We answer the question by justifying these processes to be ionization-induced multiwave mixing (or, in other words, the generation of combination frequencies) when considering ionizing two-color laser pulses with *various frequency ratios*. As shown below, the main differences between the ionization-induced multiwave mixing and well-known Kerr-like wave mixing caused by a nonlinear response of bound charges in neutrals [9,13,14] originate from the essentially high-order character of nonlinear ionization and the associated strong nonlinear dispersion. Therefore, ionization-induced wave mixing is an almost unique example of when the high-order nonlinear effects (multiwave mixing) dominate over the low-order ones (three- and four-wave mixing) in a natural way, i.e., without special deliberate compensation of low-order processes, and may be of practical interest *per se*. The number of mixed waves is determined by the effective exponent of the ionization rate as a function of the electric field strength. This number, which is typically large, is a key parameter that is related to intrinsic properties of ionized particles, parameters of a laser pulse, and features of generated radiation. This may open new ways for studying ionization dynamics as well as determining parameters of ionizing laser pulses. The concept of nonlinear ionization-induced multiwave mixing should also allow one to design schemes and methods for the generation of radiation in the THz and other frequency ranges in a sensible and targeted way.

Semiclassical and quantum-mechanical approaches.—As was discussed in previous works [11,15–21], the energy of a generated THz pulse increases with the square of the zero-frequency plasma-current density or, in other words, of the residual current density (RCD) that is left in the plasma after the ultrashort laser pulse has passed. Here we use semiclassical and *ab initio* quantum-mechanical approaches to calculate this RCD, assuming single ionization of an atom. The former approach is relevant for the tunnel ionization regime (when the electron ponderomotive energy is much greater than the ionization potential) and employs the equations for plasma density N and free-electron current density $\mathbf{j}$ [9,11,12,15–17,19–21]:

$$\frac{\partial N}{\partial t} = (N_g - N)w(E), \tag{1}$$

$$\frac{\partial \mathbf{j}}{\partial t} = \frac{e^2 N}{m}\mathbf{E}. \tag{2}$$

Here N_g is the initial density of neutral particles; e and m are the electron charge and mass, respectively; $w(E)$ is the ionization probability per unit time in the electric field of absolute value $E = |\mathbf{E}|$; and $\mathbf{E}$ is the ionizing laser field at the time moment t. The laser electric field is linearly polarized along the x axis and contains two one-color components centered at frequencies ω_0 and ω_1 with slow envelopes $A_0(t) \geq 0$ and $A_1(t) \geq 0$, respectively:

$$\mathbf{E}(t) = [A_0(t)\cos\omega_0 t + A_1(t)\cos(\omega_1 t + \varphi)]\hat{\mathbf{x}},$$

where φ is the phase shift between the components. In the calculations below, we choose the Gaussian profile for the envelopes $A_{0,1} = \mathcal{E}_{0,1}\exp(-t^2/2\tau^2)$ and the empirical formula [22]

$$w(E) = (4\omega_a E_a/E)\exp(-2E_a/3E - 12E/E_a), \tag{3}$$

which is commonly used for the tunnel ionization probability of a hydrogen atom in semiclassical models [16,19–21]. Here $\mathcal{E}_{0,1} = (8\pi I_{0,1}/c)^{1/2}$, $I_{0,1}$ are the intensities of the respective one-color components; $\tau = (4\ln 2)^{-1/2}\tau_p$, where τ_p is the laser pulse duration (intensity full width at half maximum); c is the speed of light; $\omega_a = 4.13 \times 10^{16}$ s^{-1} and $E_a = 5.14 \times 10^9$ V/cm are the atomic units of frequency and electric field, respectively. The quantum-mechanical approach is much more laborious than the semiclassical one but allows calculating the RCD in a significantly wider laser intensity range. The approach is based on the numerical integration of the three-dimensional time-dependent Schrödinger equation (3D TDSE) for an electron acted upon by the Coulomb field of the hydrogen nucleus and the laser field $\mathbf{E}$ [17,19–21]. The numerical methods used and their software implementation are detailed in Ref. [20].

Numerical simulations.—The results of calculating the RCD $j_{\mathrm{RCD}} = \lim_{t\to+\infty} j_x$ are presented in Figs. 1 and 2. If the frequency ratio $\chi = \omega_1/\omega_0$ is close to some rational fraction $a:b$ with a and b being natural and $a+b$ being a not too large odd number, the dependence of the RCD on φ is approximately periodical with the period $2\pi/b$ (here and below, we assume the fraction $a:b$ to be irreducible). Figure 1 demonstrates an example of this dependence (in the inset) and shows how the maximum (over φ) absolute value of the RCD depends on χ. The obtained dependence on χ consists of several resonantlike peaks located near frequency ratios $\chi = a:b$ with a and b natural and $a+b$ odd. The peaks are narrow enough (though wider than the spectra of one-color laser components) and are narrower the greater denominator b.

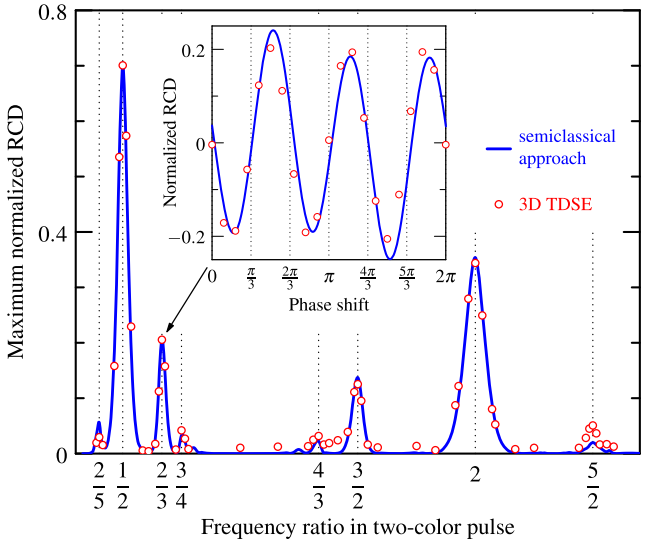

FIG. 1. Dependence of the zero-frequency (residual) current density (RCD) on the frequency ratio $\chi = \omega_1/\omega_0$ in the ionizing two-color laser pulse. The maximum (over the phase shift φ in the two-color laser pulse) absolute values of the RCD normalized to ev_aN_g are plotted ($v_a = eE_a/m\omega_a = 2.19 \times 10^8$ cm/s is the atomic unit of velocity). The frequency ω_0 is fixed and corresponds to a wavelength of 800 nm. The intensities of the one-color laser components are $I_0 = I_1 = 10^{14}$ W/cm^2, and the laser pulse duration is $\tau_p = 50$ fs. The inset illustrates the dependence of the RCD on the phase shift φ at $\chi = 2:3$. The data are obtained from a numerical solution of the 3D TDSE (circles) and semiclassical equations (1)–(3) (solid lines).

Such frequency ratios χ typically originate from synchronism conditions when rectification due to the high-order wave mixing in a centrosymmetric medium is considered. The peak widths are also consistent with synchronism bandwidths for essentially high-order wave mixing. However, contrary to what one might expect from a theory of usual high-order Kerr-like wave mixing, the peak magnitudes are inversely proportional (as the oscillatory current density is) to the laser frequencies when their ratio is fixed (it can be seen from a comparison of the peaks at $\chi = 1:2$ and $2:1$ or at $\chi = 2:3$ and $\chi = 3:2$). That is consistent with the results of Ref. [12], where the wavelength scaling of THz currents and their radiation was studied in detail. The other difference from the standard wave mixing models is that the peaks with $a+b$ larger become more pronounced at smaller intensities. At intensities of the order of 10^{13}–10^{14} W/cm^2 (such values are common to the laser filaments with the intensity clamp [14,23]), the RCD peaks at $\chi = 2:3$, $3:4$, and others may be of similar magnitude as the peaks at the commonly used frequency ratio $\chi = 1:2$. It can be seen from Fig. 2, which shows the dependence of the maximum (over φ) RCD on intensities $I_0 = I_1$ at various χ. At $I_0 = I_1 = 3\times10^{13}$W/cm^2, the ratios of the RCD at $\chi = 2:3$ and $3:4$, respectively, to the RCD at $\chi = 1:2$ are approximately equal to 0.5 and 0.2.

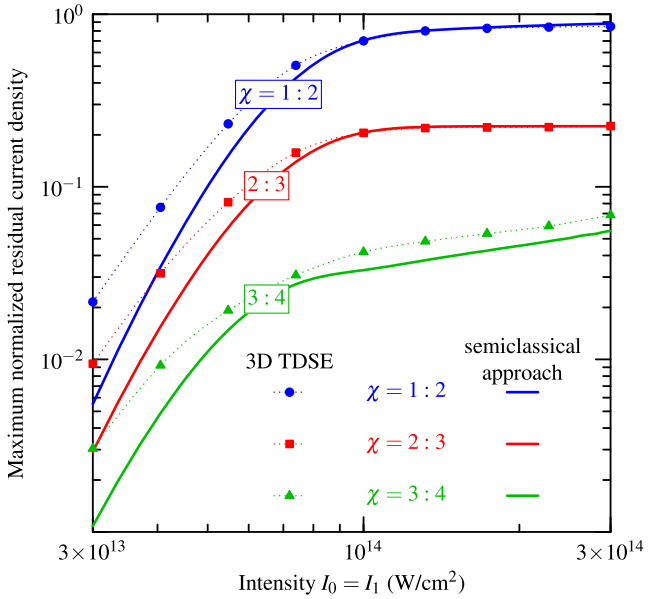

FIG. 2. The dependences of the maximum normalized RCD on intensities $I_0 = I_1$ of one-color laser components at various frequency ratios $\chi = \omega_1/\omega_0$ (see the values at the curves). The other parameters are as in Fig. 1. The solid lines represent the semiclassical numerical results, and the markers connected through by dotted lines picture the quantum-mechanical ones.

These ratios decrease with the laser pulse intensity, while the RCD increases quickly. Its growth saturates at intensities of about 10^{14} W/cm^2 due to neutral depletion. The relative strength of different peaks is also almost independent of the intensity in this case. For example, the ratio between the RCD at $\chi = 2:3$ and $\chi = 1:2$ is always greater than 0.25 for $I_1 = I_0$ and is approximately equal to that value when the neutrals are depleted. So, the presence of well-defined values for the ratios of THz yields at different peaks may provide a basis for the direct identification of ionization-induced wave mixing in experiments.

As seen from Figs. 1 and 2, the numerical results obtained from the 3D TDSE and Eqs. (1)–(3) are in quantitative agreement at laser intensities above 10^{14} W/cm^2 when the tunnel ionization regime takes place. Even though the semiclassical model is expected to give an inaccurate value for the phase shift optimal for RCD excitation [19,21], this inaccuracy does not affect the maximum absolute value of the RCD. The slight discrepancies for peaks with large $a + b$ are caused by the inexactness of the formula (3) for ionization probability. As will be clear from the analytical theory below, to describe the peaks with large $a + b$ correctly, the formula should precisely characterize not only the ionization probability itself, but also its high-order derivatives with respect to the ionizing field. At lower intensities when the ionization is beyond the tunnel regime, the semiclassical approach understates the RCD, which is a common issue [17,20]. But even so, the semiclassical model still describes the RCD qualitatively well; in particular, it correctly represents the positions and relative strengths of the resonantlike peaks in the dependence of the RCD on χ. In what follows, we develop the analytical theory of ionization-induced multiwave mixing and associated RCD excitation for the laser parameter range where the semiclassical approach is relevant.

Analytical theory.—To understand the origin of the found dependences and reinforce them, we derive analytical formulas from Eqs. (1) and (2). For that, we consider one of the one-color laser components as a perturbation and take the maximum of its envelope as a small parameter. Therefore, Taylor's approximation for ionization probability can be used:

$$w(E) \approx w(|E_0|) + E_1 w'(|E_0|)\mathrm{sgn}E_0 + \frac{E_1^2 w''(|E_0|)}{2} + \cdots, \tag{4}$$

where $E_0 = A_0(t)\cos\omega_0 t$ is the ionizing field in the zeroth order of the perturbation theory; $E_1 = A_1(t)\cos(\omega_1 t + \varphi)$ is the first-order correction to this field; and the prime means the derivative with respect to the function argument. For the sake of simplicity, we assume the ionization degree to be small, $N \ll N_g$. This allows us to write out Eq. (1) as $dN/dt = N_g w(E)$. By substituting Eq. (4) into the last equation and Eq. (2), one gets equations for the contributions N_k and j_k, respectively, to the plasma and current densities in the kth order of the perturbation theory:

$$N \approx N_0 + N_1 + \cdots; \qquad \mathbf{j} \approx (j_0 + j_1 + \cdots)\hat{\mathbf{x}};$$

$$\frac{dN_k}{dt} = \frac{N_g}{k!}(E_1\mathrm{sgn}E_0)^k w^{(k)}(|E_0|); \tag{5}$$

$$\frac{dj_0}{dt} = \frac{e^2}{m}N_0 E_0; \tag{6}$$

$$\frac{dj_k}{dt} = \frac{e^2}{m}(N_k E_0 + N_{k-1}E_1), \qquad k > 0, \tag{7}$$

where the superscript (k) denotes the derivative of the kth order with respect to the argument. One can obtain the kth-order contribution $j_{\mathrm{RCD}k}$ to the RCD from Eqs. (6) and (7) by integrating them over time: $j_{\mathrm{RCD}k} = \int_{-\infty}^{\infty}(dj_k/dt)dt$.

We consider the two-color pulses having frequency ratio χ close to $a:b$ with $a + b$ odd so that $b\omega_1 = a\omega_0 + \Delta\omega$, where $\Delta\omega \ll \omega_{0,1}$ is the detuning frequency. The pulses are presumed to be long enough (multicycle) and have slow enough envelopes $A_{0,1}(t)$ so the ionization lasts at least several field periods. The function $w(E)$ is commonly strong, and its effective exponent is $n_0 = w'(E)E/w(E) \gg 1$ [particularly, for $w(E)$ given by Eq. (3) and $I_0 = 10^{14}$ W/cm^2, $n_0 \approx 11$]. Furthermore, we assume the derivatives $w^{(k)}(E)$ corresponding to the considered orders of the perturbation theory also to be strong; the respective exponents are $n_k = w^{(k+1)}(E)E/w^{(k)}(E) \gg 1$. Normally, if $k \ll n_0$, then the last condition holds automatically, and $n_k \approx n_0 \gg 1$.

In order to catch the time dependence of N_k, we note that the right-hand side of (5) is a product of two factors: $F_k(t) = w^{(k)}(|E_0|)\mathrm{sgn}^k E_0$ and $G_k(t) = (N_g A_1^k/k!)\cos^k(\omega_1 t+\varphi)$. The first factor is a sum of even (for k even) or odd (for k odd) harmonics of the frequency ω_0 with slow amplitudes: $F_k \approx W_{k0}(t)+W_{k2}(t)\cos 2\omega_0 t+\cdots$ or $F_k \approx W_{k1}(t)\cos\omega_0 t+W_{k3}(t)\cos 3\omega_0 t+\cdots$. One can assess the slow amplitudes $W_{ks}(t)$ using the Laplace method (similarly to what was done in Ref. [11]):

$$W_{ks}(t) = \frac{\omega_0}{\pi}\int_{t-\pi/\omega_0}^{t+\pi/\omega_0} F_k(t')\cos s\omega_0 t' dt' \approx \frac{w^{(k)}(A_0)}{\sqrt{2\pi n_k}}\exp\left(-\frac{s^2}{2n_k}\right).$$

The second factor is likewise a superposition of even (for k even) or odd (for k odd) harmonics of ω_1, and the maximum possible harmonic number is k. Therefore, in accordance with Eqs. (5) and (7), the contributions N_k and dj_k/dt consist of harmonics with slow amplitudes at certain combination frequencies of ω_0 and ω_1. In the integral over time for $j_{\mathrm{RCD}k}$, all harmonics in dj_k/dt at high frequencies vanish due to the averaging, and only harmonics with frequencies $p\Delta\omega = pb\omega_1 - pa\omega_0$ and a not too large natural p contribute to $j_{\mathrm{RCD}k}$. The minimal order of the perturbation theory that gives a nonzero RCD is b, and the RCD is determined by $j_{\mathrm{RCD}b}$.

In accordance with Eq. (7), one should find amplitudes of harmonics within N_b at frequencies $\omega_0 \pm \Delta\omega$ and amplitudes of harmonics within N_{b-1} at frequencies $\omega_1 \pm \Delta\omega$ in order to evaluate $j_{\mathrm{RCD}b}$. By substituting these harmonics into Eq. (7) and simplifying the resulting equation under the assumption $a+b \ll n_0$ for small $\Delta\omega$, we find

$$\frac{dj_b}{dt} \approx j_{\mathrm{osc}} g\bar{w}(A_0)\left(\frac{n_0 A_1}{2A_0}\right)^b \sin(\Delta\omega t + b\varphi), \qquad (8)$$

where $j_{\mathrm{osc}} = e^2 N_g A_0/m\omega_0$ is the oscillatory current density induced by the one-color (unperturbed) laser field in a plasma of density N_g; $\bar{w}(A_0) = (2/\pi n_0)^{1/2} w(A_0)$ is the period-averaged ionization probability; and g is the factor that depends only on the frequency ratio ω_1/ω_0 and the exponent n_0 of function $w(E)$:

$$g = \frac{2}{b!}\left(\frac{a^2-b^2}{n_0 a} - \frac{\Delta\omega}{\omega_0}\right)\exp\left(-\frac{a^2}{2n_0}\right). \qquad (9)$$

By integrating Eq. (8) over time and using the Laplace method, we obtain the RCD

$$\mathbf{j}_{\mathrm{RCD}} = j_{\mathrm{osc}} g\sigma \exp\left(-\frac{\Delta\omega^2\tau_i^2}{2}\right)\left(\frac{n_0\mathcal{E}_1}{2\mathcal{E}_0}\right)^b \times \sin(b\varphi - \Delta\omega t_0)\hat{\mathbf{x}}, \qquad (10)$$

where t_0 is the time of maximum $A_0(t)$; $\mathcal{E}_{0,1} = A_{0,1}(t_0)$ are the envelope values at this time; $\tau_i = \tau/n_0^{1/2} \ll \tau$ is the ionization duration that characterizes the time width of the factor $\bar{w}(A_0)$ in Eq. (8) and indicates how long the ionization lasts; $\tau = [-A_0/(d^2A_0/dt^2)]^{1/2}|_{t=t_0}$ is the duration of the unperturbed laser field; $\sigma = (2\pi)^{1/2}\tau_i\bar{w}(\mathcal{E}_0)$ is the ionization degree reached at the end of the laser pulse; and the exponent n_0 also corresponds to the time t_0.

The assumption $N \ll N_g$ used in the derivation is not critical. Following the procedure in Ref. [20], one can generalize the obtained formula for greater ionization degrees. The resulting equation for the RCD has the same form (10), but the values of σ and τ_i should be adjusted, and the moment of the maximum averaged ionization rate should be taken as t_0. With these adjustments, the formula (10) can describe the RCD even at high enough intensities when $\sigma \sim 1$.

The obtained closed-form formula (10) describes well virtually all effects found in the numerical simulations. Particularly, it describes the peak positions and widths (and the peak magnitudes as dependent on ω_0) as well as the qualitative dependence on the total laser intensity. The key parameter that determines these dependences is the effective exponent n_0 of the function $w(E)$. In the ionization-induced wave mixing under consideration, n_0 plays a similar role to that of the number of mixed waves (wave mixing order) in the common high-order wave mixing. Indeed, for example, the duration of a generated nonlinear current [described by Eq. (8)] is equal to the ionization duration τ_i, which is $n_0^{1/2}$ times shorter that the laser pulse duration just as the duration of the nonlinear polarization from n_0-wave mixing is. Just like in common high-order wave mixing, exactly this parameter determines the maximum values a and b where peaks are prominent and the ionization-induced wave mixing still takes place: $a+b \lesssim n_0$. And for small I_1, the dependences of the RCD on I_1 present power laws with the exponents $b/2$ and are similar to those coming from the common high-order wave mixing models for a zero-frequency nonlinear polarization response.

In fact, many aspects of the effect under consideration can be described in the framework of the phenomenological multiwave mixing model. However, there are substantial differences between the ionization-induced wave mixing and the common models of multiwave mixing. The following distinctions are apparent from our analytical and numerical results: (i) The number n_0 of mixed waves drastically depends on the laser intensity; (ii) the effect is strongly modified at high enough intensities due to the neutral depletion; (iii) there is an asymmetry with respect to the sign of the detuning $\Delta\omega$. The last is evident from Eq. (9): Replacing $\Delta\omega$ with $-\Delta\omega$ generally changes the value g, which is uncommon for typical high-order nonlinearities such as the instantaneous high-order Kerr effect.

This asymmetry is a manifestation of the nonlinear dispersion associated with ionization and may be useful in determining the origin of generated THz radiation (or radiation in other frequency ranges) in experiments with two-color laser pulses. The identification of this origin presents one of the fundamental questions in the theory of two-color laser-plasma THz generation [9].

The other example of a benefit from the presented theory is the following. According to Eq. (10), the peak widths are equal to $1/\tau_i b$, and the 3D TDSE numerical simulations support this relation for a wide enough parameter range. This particular fact presents a method to reconstruct the ionization duration and corresponding exponents n_0 in a rather direct and simple way from measurements of the peak widths in the dependence of the THz yield on the detuning frequency.

Conclusions.—We examine the new type of nonlinear optical interaction that occurs in a plasma created by two-color ultrashort laser pulse with an arbitrary frequency ratio of its one-color components and can be identified as the ionization-induced multiwave mixing. Based on *ab initio* quantum-mechanical and semiclassical calculations, we show that the excitation of the RCD of free electrons responsible for THz generation is attributed to this wave mixing. The main features of this wave mixing are defined by the intrinsic nonlinear properties of the ionized particles. Particularly, the number of mixed waves is determined by the effective exponent of the ionization rate as a function of the ionizing field strength. The dependences of the maximum (over the phase shift between one-color components) RCD on the frequency ratio consist of resonantlike peaks at frequency ratios corresponding to rational fractions with a not so big odd sum of the numerator and the denominator (fractions such as $1:2$, $2:3$, $3:4$, $2:5$, etc.). The magnitudes of different peaks can be comparable when the two laser components have close intensities, and the laser-plasma THz generation with the two-color pulses of uncommon frequency ratios may be effective enough.

Our closed-form analytical formula (10) supports the above conclusions and reveals the similarities and differences between the ionization-induced wave mixing under consideration and the common wave mixing associated with the Kerr-like nonlinear response of bound charges. We envisage that the identification of these similarities and differences will contribute to (i) the development of a whole new class of schemes for radiation generation at laser combination frequencies (which may occur in various frequency ranges), (ii) the determination of wave mixing type in particular experiments, and (iii) the development of new methods for probing the ionization dynamics, particularly, for the determination of ionization durations.

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