Collisional dynamics in laser-induced plasmas: evidence for electron-impact excitation

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Abstract: We have experimentally investigated the collisional dynamics in femtosecond-laser-induced plasmas and presented the evidence for electron-impact excitation through enhanced high-order harmonic (HH) generation. The measurements were carried out by using an elliptically polarized pump pulse to induce the underdense plasmas and by using a time-delayed linearly polarized probe pulse to drive the HH generation from the plasmas. We found that the rise time of this enhanced HH generation was insensitive to the ellipticity degree (ED) of pump pulse but sensitive to its laser intensity (LI). With further comparison between physical scenarios and qualitative analysis, we demonstrated that the atomic excitation causing the HH enhancement should be attributed to the electron-impact excitation, i.e., the excitation from the collision between neutral atoms and electrons during the lifetime of the underdense plasma.

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OCIS codes: (020.2649) Strong field laser physics; (190.2620) Harmonic generation and mixing; (320.2250) Femtosecond phenomena; (320.7110) Ultrafast nonlinear optics.

References and links


1. Introduction

High-order harmonic (HH) generation is a universal response of matter to strong femtosecond laser fields, and has always been a hot topic of strong field physics and attosecond science [1,2]. Materials for the HH generation have evolved from initially simple gaseous atom [3,4] and molecule [5–10] to recently complicated solid [11–14], underdense plasma [15–17] and even filament [18,19]. In general, HHs are emitted during the recombination of an accelerated electron with a core [20,21], through which information about structure and dynamics of the target is encoded in the HH amplitude and phase with an unprecedented spatial and temporal resolution. Hence, the HH spectroscopy has been widely used in the study of ultrafast electron dynamics in atoms [22,23], chemical reaction and electron dynamics in molecules [24–26], and electron dynamics in solids [12]. Here, we extend the HH spectroscopy to femtosecond laser-induced plasmas, and present the experimental evidences for electron-impact excitation during the lifetime of underdense plasmas. Recent pump-probe experiment [15] has shown that the HH yield in excited Argons exhibits high conversion efficiency with a certain elliptical polarization of pump pulse. The generation of excited atoms was attributed to frustrated tunneling ionization (FTI) [27–29], and thus the enhanced HH yield from such excitation should be sensitive to the ellipticity degree (ED) of pump pulse. However, in our experiment, we found that such enhanced HH yield is not sensitive to the pump ED, and a more reasonable electron-impact excitation mechanism is proposed [30,31].

In this work, an elliptically polarized laser is introduced as a pump pulse to induce an underdense plasma in an argon-filled cell, while a linearly polarized laser is employed as a probe pulse to drive the HH emission. We have experimentally measured the HH yield as a function of time delay between the pump and probe pulses, and compared the dependence of the HH enhancements on the pump ED with that on the pump laser intensity (LI). It is found that the rise time of such enhancement is more sensitive to both the pump LI and the gas pressure than to the pump ED. These features are closely related to and well explained by the electron-impact excitation mechanism, i.e., the excitation from the collision between neutral atoms and electrons during the lifetime of this laser-induced plasma. Moreover, through the comparison between physical scenarios and qualitative analysis, such a mechanism of collisional excitation has been further confirmed.

2. Experiment

A commercial Ti:sapphire femtosecond laser (Coherent, Inc.) is used to produce 8 mJ laser pulses at 800 nm center wavelength with a pulse duration of 45 fs at a repetition rate of 1 kHz. The output pulse of the laser system is split into two beams, where one is used as a pump pulse for preparing an underdense plasma and the other as a probe pulse for driving HHs. The two beams are focused by convex lenses with a focal length of 500 mm on the center of a 25-mm long and argon-filled gas cell located in a high vacuum interaction chamber. Finally, an aluminum foil with a thickness of 500 nm is used to block the residual driving laser, and the generated HHs are detected by a homemade flat-field grating spectrometer equipped with a soft x-ray CCD (Princeton Instruments, SX 400). In order to remove the contribution of HHs from the pump pulse, the elliptical polarization of certain value is introduced by using a quarter-wave plate for the pump pulse to suppress the HH generation. It is ensured that the collected HHs are primarily generated by the linearly polarized probe pulse. Moreover, in order to distinguish the HH signals from the different atomic states, referred to as the ground-state, the excited-states and the ionized state, the moderate LI of the probe pulse is chosen to only drive HH generation from the atoms in ground / excited states in this pump-probe configuration. The normal HH signal is generated from the neutral atoms. The enhanced HH signal is generated from the excited atoms [15,32].
Whereas, the ionized atoms cannot emit HH signals. Hence, the probe LI is kept constant about $2 \times 10^{14}$ W/cm². The pump LI is tuned from 0 to $5 \times 10^{14}$ W/cm² by using a half-wave plate and a high extinction film polarizer. In order to obtain the optimal HH signals, an aperture-iris diaphragm is used to improve the beam profile and the pump focus position is moved around the center of the gas cell. To reduce the effect of phase matching, a very low gas pressure of about 20 Torr is chosen. The experimental LI can be estimated by $I = \frac{2E}{\pi \tau w^2}$, where $E$ is the measured laser energy, $\tau$ is the measured pulse duration, and $w$ is the $1/e^2$ beam radius.

3. Results and discussion

![Fig. 1. Experimentally measured harmonic yield as a function of time delay between the pump and probe pulses. (a)-(c) For different pump LIs: (a) $2.0 \times 10^{14}$, (b) $2.5 \times 10^{14}$, and (c) $3.5 \times 10^{14}$ W/cm², with the same ED of 0.35. (d)-(f) For different pump EDs: (d) 0.25, (e) 0.45, and (f) 0.60, with the same LI of $3.5 \times 10^{14}$ W/cm².]

Using the above experimental scheme, the HH yield as a function of the time delay between the pump and probe pulses is measured. In Figs. 1(a)-1(c), the results for different pump LIs but a fixed pump ED of 0.35 are presented. While in Figs. 1(d)-1(f), we present the delay-dependent harmonic spectra for different pump EDs with a fixed pump LI of $3.5 \times 10^{14}$ W/cm². From Figs. 1(a) to 1(c), one can see that the location and extent of the time-dependent enhancement change obviously when the pump LI increases. However, they are almost invariant when the pump ED changes, as shown in Figs. 1(d)-1(f). For quantitative comparison, we extract the HH yield from Fig. 1 by summing over all the harmonics, and present the integrated HH yield in Fig. 2(a) for the case of varying LI and in Fig. 2(b) for the case of varying ED, respectively. One can see that the enhancement of the HH signal appears immediately and increases gradually after the pumping for both two cases. In the case of varying LI shown in Fig. 2(a), the rise time of the significant enhancement are estimated to be 25, 18, and 5.5 ps for the LIs of $2.0 \times 10^{14}$, $2.5 \times 10^{14}$, and $3.5 \times 10^{14}$ W/cm², respectively. While in the case of varying ED shown in Fig. 2(b), they are 5, 6, and 7 ps for the EDs of 0.25, 0.45 and 0.60, respectively. Here, the rise time is defined as the time when significant enhancement is observed. Through the above comparisons, one can obviously see that the rise time of the HH enhancement resulting from the excited atoms are more sensitive to the variation of the LI than to the variation of the ED. This strong dependence of the HH enhancement on the pump LI is quite similar to the dependence of the kinetic energy of the released electrons on the pump pulse, i.e. the kinetic energy of the released electron is mainly
determined by the pump LI. This similarity suggests that the atomic excitation causing the HH enhancement is mainly resulted from the electron-impact excitation rather than the FTI [33]. The rise time of the HH enhancement corresponds to the mean collision time of the electron-impact excitation when the significant excitation of atom occurs. A further discussion will be presented in the following. As shown in Fig. 2, the integrated HH yields also present some oscillations, of which the origin is an open question.

![Fig. 2. HH yield obtained by summing over all the harmonics shown in Fig. 1. (a) from Fig. 1(a)-1(c), the rise time of the enhancements are 25, 18 and 5.5 ps for the LIs of 2.0 × 10^{14}, 2.5 × 10^{14}, and 3.5 × 10^{14} W/cm², respectively; (b) from Fig. 1(d)-1(f), the rise time of the enhancements are 5, 6, and 7 ps for the EDs of 0.25, 0.45 and 0.60, respectively.](image)

In Ref [15], the excitation was attributed to the FTI when the gas is exposed to the elliptically polarized laser. According to the FTI [27], the HH enhancements from such excitation should occur during the short pulse overlap and should be sensitive to the pump ED. However, our experimental results show that the HH enhancements occur during the long lifetime of plasma (i.e., continuing for a long time after the delay of 0 ps) and are insensitive to the variation of the pump ED. Therefore, the FTI mechanism cannot explain the observed dependence of the HH enhancement on the pump pulse. We proposed that the electron-impact excitation mechanism may play an important role in our experiment. To analysis the collisional dynamics in these plasmas, we further measured the HH yield for more pump LIs of 1, 2, 3.5, and 5 × 10^{14} W/cm². The results are shown in Figs. 3(a)-3(d). One can see that there are three cases. Case 1: at a low pump LI, there is no change for HH signal as shown in Fig. 3(a). Case 2: at a moderate pump LI, the harmonic enhancements are observed at certain delays, as shown in Figs. 3(b)-3(c). Case 3: at a strong pump LI, the HH emission is totally suppressed for all the delays as shown in Fig. 3(d). The schematic diagrams for these physical scenarios are shown in Fig. 3(e). For the low pump LI in Case 1, the pump LI is insufficient to generate gas plasma, hence only the normal harmonics are observed. For the high pump LI in Case 3, the target is totally ionized by the pump pulse and no HH signal could be emitted because the probe LI is not strong enough to generate HHS from the ionized states. For the moderate pump LI in Case 2, the scenario becomes complicated. The rise times and the magnitudes of the enhancements depend on the pump LI, as shown by the Case 2A and Case 2B, i.e., the higher pump LI is used, the earlier and stronger enhancement of the HH signal is observed. This phenomenon is consistent with the electron-atom collision mechanism.

According to thermodynamics, the mean collision time between electrons and atoms can be estimated by

\[
\tau_{\text{e-atom}} = \frac{\lambda_{\text{e-atom}}}{\nu_e} = \frac{(n\sigma)^{-1}}{2E_k/m_e} = \frac{k_B T m^{1/2}}{\sqrt{2\pi \eta P\sigma E_k^{1/2}}} \tag{1}
\]
where $v_e = \sqrt{\frac{2E_k}{m_e}}$ is the velocity of released electron, $E_k$ is the kinetic energy of released electron (the maximum kinetic energies $E_k$ can be estimated by $3.17U_p$), $m_e$ is the mass of electron, $n = \frac{\eta P}{k_B T}$ is the number density of neutral atoms, $k_B$ is the Boltzmann constant, $T$ is the atom temperature ($T = 300$ K), $\eta$ is the population of the remaining neutrals and can be estimated by ADK theory [34], $P$ is the gas pressure, and $\sigma$ is the total cross section of argon from experimental data [35,36].

From Eq. (1), one notes that the mean collision time between the remaining neutrals and the released electrons, corresponding to the rise time of the HH enhancements, are inversely proportional to both the kinetic energy $E_k$ ($E_k$ is directly relevant to LI) and the gas pressure $P$. Indeed, we have experimentally observed these two physical predictions as shown in Fig. 4(a)-4(b), together with the theoretical simulation from Eq. (1) as shown in Fig. 4(c)-4(d).

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Figure 4(a) is the reproduction of Fig. 2(a) for comparison with Fig. 4(c). In Fig. 4(c), the solid-red, dashed-green, and dotted-blue curves represent the neutral population as a function of the mean collision time for the pump pulses of $3.5 \times 10^{14}$, $2.5 \times 10^{14}$, and $2 \times 10^{14}$ W/cm², respectively. The mean time of the electron-neutral collision is directly calculated by Eq. (1) as a function of neutral population by using the free-electron energies of 3, 5, and 15 eV. The kinetic energies of 3, 5, and 15 eV are the fitting results through using the mean collision time of 25, 18, and 5.5 ps by Eq. (1). It is clearly shown that the higher pump LI is used, the earlier and stronger enhancement of the HH signal is observed. As discussed, the mean collision time for the electron-impact excitation is consistent with the rise time of the harmonic
enhancement. By applying this relation, the population of the remaining neutrals can be extracted from the curve representing the neutral population as a function of the mean collision time, or equivalently the rise time. This result is shown in Figs. 4(a) and 4(c), and the populations of the remaining neutrals can be read from the curves as 50%, 60%, and 95% for the pump LIs of $3.5 \times 10^{14}$ W/cm², $2.5 \times 10^{14}$ W/cm² and $2 \times 10^{14}$ W/cm², respectively. These values are close to those calculated based on the ADK theory [34].

In Fig. 4(b) with the enlarged view of the rising part, we further measured the delay-dependent HH signal for various gas pressures with a fixed pump LI of $3.5 \times 10^{14}$ W/cm². One can see that the rise time of the HH enhancement are observed at the delays of 2, 5.5, and 7 ps for the cases with gas pressures of 50, 20, 15 Torr, respectively. In other words, the higher the gas pressure is, the earlier the HH enhancement occurs. This phenomenon also supports the mechanism of electron-impact excitation.

In the following, we demonstrate how to extract the kinetic energy of the electron involving in the collision by using the relevance of the collision time to the rise time of the harmonic enhancement. After obtaining the rise time of the harmonic enhancement, we substitute the estimated neutral population (50%) for the pump LI of $3.5 \times 10^{14}$ W/cm² into Eq. (1) to obtain the curves of the kinetic energy versus the mean collision time. The results are shown in Fig. 4(d) by the solid-red, the dashed-green, and the dotted-blue curves for the cases with gas pressures of 50, 20, 15 Torr, respectively. Finally, applying the consistence between the rise time of the harmonic enhancement and the collision time, the kinetic energy responsible for the collision can be extracted. As shown in Figs. 4(b) and 4(d), the kinetic energies are around 15 eV for all the cases with different gas pressures. This is not surprising, because the kinetic energy of the released electron is determined by the pump laser pulse. Meanwhile, the pump laser pulse is the same for the three cases with different gas pressures. From the above discussion, one can see that by applying the relevance of the rise time of the harmonic enhancement to the mean collision time for the electron-impact excitation, the
population of the neutral in gas plasma and the kinetic energy of the released electron involving in the collision can be roughly estimated.

Therefore, the collisional dynamics in the laser-induced plasmas can be understood as follows: when the plasma is immediately produced after the neutral atoms being illuminated by the pump pulse, the excited atoms are first generated by the collision of the remaining neutrals with the free electrons on the time scale of several picoseconds. Then they are depleted by the further collisions of the excited atoms with the later arriving electrons on the time scale of dozens of picoseconds, and this depletion time is consistent with the simulation of Ref [15]. Furthermore, there should be some complicated spatial dynamics (such as phase-matching effect and probe-defocusing effect) to affect the HH signals. These effects may affect the amplitudes of the delay-dependent HH signals, but they have little effect on the time-delay-dependent structure observed here because these dynamics mainly occur on the femtosecond time scale, much shorter than the picosecond time scale of the present work. Our simple theoretical simulation captures the major physics of the electron-impact excitation process, and gives a qualitative explanation. However, a more comprehensive theory, considering the propagating and phase-matching effects, is still necessary for fully account for the underlying physics.

4. Conclusion

In conclusion, the collisional dynamics and the physical scenarios in femtosecond-laser-induced plasmas have been investigated by using the HH spectroscopy. The results show that the rise time of the HH enhancement is sensitive to the pump LI and the gas pressure but not to the pump ED. A further comparison including the qualitative analyzation confirm the generation of atomic excitation by the electron-atom collision.

Funding

National Natural Science Foundation of China (NSFC) (11474223, 11234004, 61504082, 11604248); Global Research Laboratory Program (2009-00439); Max Planck POSTECH/KOREA Research Initiative Program (2016K1A4A4A01922028).

Acknowledgment

This experimental work was supported by the State Key Laboratory of High Field Laser Physics of the Shanghai Institute of Optics and Fine Mechanics, CAS.