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Insights from Keldysh theory to plasma electron density in liquid water under excitation wavelength scaling

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Abstract
The study of plasma generation in liquids is relevant for many applications, especially for increasing the efficiency of terahertz radiation generation. This work investigates the relationship between the laser excitation wavelength and the plasma electron density in liquid water in the near-infrared spectral range. Using numerical simulation methods based on the Keldysh theory, patterns of changes in the ionization rate and changes in the plasma electron density depending on the excitation wavelength are analyzed. The results show the mutual influence of above-threshold ionization and tunneling effects when the Keldysh parameter is close to one. A decrease in plasma electron density with increasing excitation wavelength has been shown. However, in certain wavelength ranges a local increase in plasma electron density was observed. The theoretical results obtained are consistent with the experimental data of other scientific groups. This theoretical study provides valuable information on the modulation of plasma electron density by changing laser excitation wavelengths, which is important for increasing the efficiency of terahertz radiation generation.

Keywords
plasma, Keldysh theory, ionization, liquid, wavelength, plasma-based THz generation, plasma electron density

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УДК 533.9

Оценка плотности плазмы в воде на основе теории Келдыша при изменении длины волны накачки

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Аннотация
Введение. Исследование генерации плазмы в жидкостях актуально для многих областей применения. Главным образом данное исследование применяется для повышения эффективности генерации терагерцового излучения. В работе исследуется взаимосвязь между длиной волны лазерной накачки и плотностью плазмы для воды в

ближнем инфракрасном диапазоне спектра. **Метод.** Методами численного моделирования, основанного на теории Келдыша, выполнен анализ закономерности изменений скорости ионизации и плотности плазмы от длины волны накачки. **Основные результаты.** Полученные результаты показали наличие взаимного влияния сверхпороговой ионизации и туннельных эффектов, преимущественно в случае, когда параметр Келдыша близок к единице. Показано снижение плотности плазмы при увеличении длины волны излучения накачки. Однако при этом было выявлено локальное увеличение плотности плазмы в определенных диапазонах длин волн. **Обсуждение.** Полученные теоретические результаты согласуются с экспериментальными данными подобных научных работ. В результате теоретического исследования получена важная информация о модуляции плотности плазмы с помощью изменения длин волн лазерного возбуждения, что имеет значение для повышения эффективности генерации терагерцового излучения.

Ключевые слова

плазма, теория Келдыша, ионизация, жидкость, длина волны накачки, генерация ТГц излучения из плазмы, плотность плазмы

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Introduction

Enhancing the efficiency of terahertz (THz) generation has become a significant goal for the scientific community, given its crucial role in various applications, including material science, radiation sources, optical coatings, and more [1, 2]. Liquids, as a dense medium, allow for more energy transfer and confinement compared to gases [3]. This can be a plus for obtaining faster and higher ionization giving plasma-based applications in liquids an advantage [4]. Plasma-based THz radiation in liquids is among the most efficient sources achieving optical-to-THz conversion efficiency of a few percent [5]. Further investigation is needed for ongoing development. Prior data on liquid-based THz sources highlights the impact of various pump parameters on THz generated emission but is limited to the central wavelength of 800 nm. However, in [6], the THz generation process was experimentally observed under long-wavelength excitation (1200–1500 nm). The results indicate that in a water liquid jet at pump pulse energy of 0.27 mJ, the THz field enhances with the excitation wavelength increasing. Additionally, a theoretical study [7] demonstrated that THz efficiency increases with the excitation wavelength in the mid-infrared region. Earlier [8] it was demonstrated that by gas ionization, at a specific focusing conditions dependent on the wavelength at 1800 nm, the efficiency of THz generation enhances by 30 times relative to the excitation wavelength of 800 nm.

Given that different excitation wavelengths result in varying plasma electron densities and frequencies, these findings have captured our attention in the context of plasma physics behind the THz generation process in liquid jets. Recent studies [9, 10] on laser-induced plasma-based THz generation, conducted in the presence of an external electrical field, have revealed a proportional relationship between plasma electron density and THz efficiency. It has been observed that the vertical component of THz radiation is directly proportional to the plasma electron density along the laser filament. This highlights the pivotal role of plasma electron density and frequency in determining the efficiency of THz generation. Therefore, further investigation into plasma electron density is imperative to deepen our understanding of this crucial aspect. Many works have already investigated the relation of plasma electron density and the wavelength of excitation. There seems to be a decreasing trend of plasma electron density when increasing the excitation wavelength [11–13].

In this study, we will investigate the plasma electron density dependency on the excitation wavelength within the Near-Infrared (NIR) range in liquid water based on the general Keldysh theory [14–16], which has revealed unexpected behavior of plasma electron density increasing for specific wavelength ranges when taking into account the Above-Threshold Ionization (ATI), differing from the overall continuously decreasing plasma electron density trend occurs with increasing excitation wavelength [11–13]. We will provide a comprehensive explanation of the obtained results and confirm them based on existing works [11, 17].

Theoretical Background

Keldysh theory predicts rates of multiphoton and tunneling ionization under strong laser fields enabling the calculation of plasma electron density. The crucial Keldysh parameter γ defining the ionization process as tunneling or multiphoton depends on the ratio between the ionization energy of the medium ion Δ, and the ponderomotive energy of the laser field U_P [16, 18].

$$\gamma = \sqrt{\frac{\Delta}{2U_P}} = \frac{\omega}{e}\frac{\sqrt{m_e\Delta}}{F}, \quad (1)$$

$$U_P = \frac{e^2F^2}{4m_e\omega^2}. \quad (2)$$

Where ω is the angular frequency of the laser field, F is the laser electric field amplitude, m_e is the electron mass, and e is the electron charge. If the ponderomotive energy which is the average kinetic energy of a free electron under the influence of the laser field surpasses the ionization energy $\gamma << 1$, electron liberation from the parent ion can occur without reliance on resonant processes, such as multiphoton absorption, in this case tunneling and over-the-barrier ionization occurs.

Otherwise, if $\Delta >> U_P$, then $\gamma >> 1$, the laser field may lack sufficient strength to expel electrons from their ions allowing multiphoton processes to potentially play the dominate role. In this context, there is a case wherein absorbing multiple photons surpasses the ionization potential by more than one photon to access the ionization continuum. This phenomenon is known as ATI which extends beyond multi-photon ionization [18]. It occurs when the ionization energy surpasses both the ponderomotive energy and photon energy $\Delta > U_P > \hbar\omega$, where $\hbar$ is reduced Planck's constant. While the Keldysh parameter aids in identifying the dominant process as multiphoton ionization when $\gamma >>1$ or tunneling ionization when $\gamma << 1$, it may not always distinguish between the two regimes, especially when γ is close to 1 where both multiphoton and tunneling ionization can occur concurrently resulting in a blend of both effects [19].

At high power intensities, liquids exhibit quasi-metallic transient optical properties and are treated within the framework of a non-parabolic band model. In this case, the widely recognized full Keldysh ionization rate W_{NP} for non-parabolic band model that accounts for both the tunneling and the multiphoton ionization is applied [14, Eq. (37)]:

$$W_{NP} = \frac{4\omega}{9\pi}\left(\frac{m_e\omega}{\gamma_1\hbar}\right)^{3/2} Q(\gamma, x) \times \times \exp\left(-\pi\langle x+1\rangle \frac{K(\gamma_1)-E(\gamma_1)}{E(\gamma_2)}\right), \quad (3)$$

guided by the respective γ values obtained.

In Eq. (3), $\gamma_1 = \gamma/\sqrt{1+\gamma^2}$ and $\gamma_2 = \gamma_1/\gamma$, ω is the angular frequency of the laser field, K and E are the complete elliptic integrals functions of the first and second kind respectively. $Q(\gamma, x)$ is a function pertaining to the structure of the spectrum which is associated with the discreteness of the absorbed photon count given by the formula [14, 15]:

$$Q(\gamma, x) = \sqrt{\frac{\pi}{2K(\gamma_2)}}\sum_{n=0}^{\infty}\left[\exp\left(-\pi n\frac{K(\gamma_1)-E(\gamma_1)}{E(\gamma_2)}\right) \times \times \Phi\left(\sqrt{\frac{\pi^2(\langle x+1\rangle - x + n)}{2K(\gamma_2)E(\gamma_2)}}\right)\right].$$

Here n is the number of absorbed photons and Φ can be defined by $\Phi(z) = \int_0^z e^{y^2-z^2}dy$, $x = \Delta^*/\hbar\omega$ where $\langle x+1\rangle$ is the minimum number of photons needed to be absorbed by the ion to ionize, and Δ^* is the effective ionization potential given by $\Delta^* = 2\Delta E(\gamma_2)/\pi\gamma_1$ [15].

The primary parameters that pertain to the medium, namely the ionization energy (Δ) and refractive index (n_0) described by the effective ionization potential formula. For low pump frequencies and a strong laser field ($\gamma << 1$), the ionization probability described by Eq. (3) simplifies to the pure tunneling effect formula given in [14, Eq. (40)]:

$$W_{NP} = \frac{2\Delta}{9\pi^2\hbar}\left(\frac{m_e\Delta}{\hbar}\right)^{3/2}\left(\frac{e\hbar F}{m_e^{1/2}\Delta^{3/2}}\right)^{5/2} \times \times \exp\left[\frac{-\pi}{2}\frac{m_e^{1/2}\Delta^{3/2}}{e\hbar F}\left(1 - \frac{1}{8}\frac{m_e\omega^2\Delta}{e^2F^2}\right)\right]. \quad (4)$$

On the other hand, in case of week laser field ($\gamma >> 1$), the ionization probability described by Eq. (3) simplifies to the pure multi-photon effect formula given in [14, Eq. (41)]:

$$W_{NP} = \frac{4\omega}{9\pi}\left(\frac{m_e\omega}{\hbar}\right)^{3/2}\Phi\left(\sqrt{2\langle\tilde{x}+1\rangle - 2\tilde{x}}\right)\left(\frac{e^2F^2}{16m_e\omega^2\Delta}\right)^{\langle\tilde{x}+1\rangle} \times \times \exp\left[2\langle\tilde{x}+1\rangle\left(1 - \frac{e^2F^2}{4m_e\omega^2\Delta}\right)\right], \quad (5)$$

$\tilde{x} = \tilde{\Delta}/\hbar\omega$ where the ionization potential in this case takes the value $\tilde{\Delta} = \Delta + (e^2F^2/4m_e\omega^2)$.

Eqs. (3)–(5) describe the average ionization probability over a time much larger than the period of the external field ($2\pi/\omega$). They represent the number of free electrons generated per unit volume per unit time. To calculate plasma electron density ρ_e, one should integrate Eqs. (3)–(5) the average ionization probability over the period during which the laser field interacts with the medium [20]. After calculating the plasma electron density ρ_e, the plasma electron frequency can be obtained by $\omega_P = \sqrt{\rho_e e^2/m_e\varepsilon_0}$ [10, 21].

Results

In our simulation, the selection of the pump parameters is predicated upon their widespread utilization in numerous experimental investigations focusing on plasma-based THz generation in liquids. We are considering the ionization in a water liquid jet excited by a femtosecond laser with a pulse energy of 300 μJ, pulse duration of 150 fs, FWHM of 100 μm, resulting in a peak power density of $I_{peak} = 2.54\cdot10^{13}$ W/cm^2.

Having the ionization potential of water in hand $\Delta = 11.67$ eV $= 1.869\cdot10^{-18}$ J, then, in the wavelength ranging from 0.8 μm to 2.2 μm, Keldysh parameter γ took the values presented Fig. 1, *a* according to Eqs. (1), (2).

Analysis of the provided data in Fig. 1, *b*, *c* suggests that the condition for ATI is satisfied since the ionization energy of liquid water $1.87\cdot10^{-18}$ J exceeds both the ponderomotive energy and photon energy values, whereas the ponderomotive energy is higher than the photon energy values in the studied wavelength range, as shown in Fig. 1, *b*, *c*, where ($\Delta > U_P > \hbar\omega$). In this case, one should conduct the simulation using Eq. (3). Fig. 2, *a* represents the plasma electron density calculated based on Eq. (3) as a function of the excitation wavelength accompanied by a miniature representation of the logarithmic scale in the upper right corner to better illustrate the decrease in values.

In Fig. 2, the plasma electron density initially registers $9.58\cdot10^{19}$ cm^{-3} at a wavelength of 0.8 μm, subsequently peaking at 0.83 μm for the minimum value of $\langle x+1\rangle = 8$, and it reaches $2.56\cdot10^{12}$ cm^{-3} at a wavelength of 2 μm.

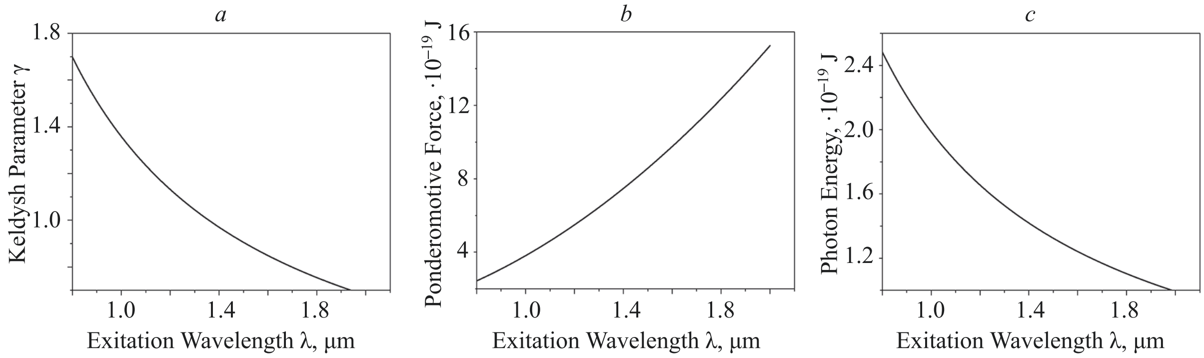

Fig. 1. Keldysh parameter γ (*a*), the ponderomotive energy of the laser field U_P (*b*), and Photon energy $\hbar\omega$ as functions of excitation wavelength (*c*)

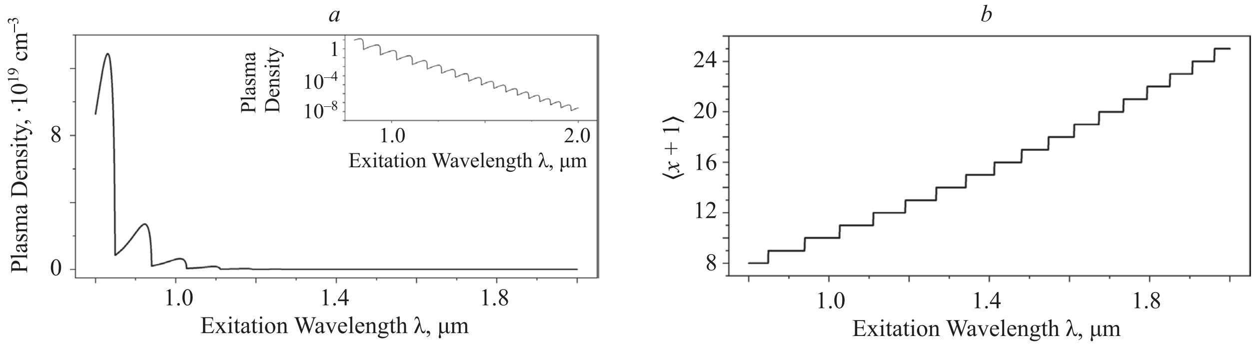

Fig. 2. Plasma electron density ρ_e as function to the excitation wavelength with a miniature representation of the logarithmic scale in the upper right corner (*a*). The dependency of $\langle x + 1\rangle$, the minimum number of photons needed to ionize the ion, on the excitation wavelength (*b*)

We observe an initial trend: for each $\langle x + 1\rangle$ considered independently, the plasma frequency increased with the excitation wavelength until reaching saturation. However, a counteracting effect occurred as the wavelength increased. Simultaneously, the minimum number of photons $\langle x + 1\rangle$ also increased leading to an overall decrease in the plasma electron density. In essence, as evident in the representation of the logarithmic scale of Fig. 2, *a*, the plasma electron density tends to decrease with an increase in excitation wavelength. This gradual decreasing trend can be elucidated by the relationship between excitation wavelength and photon energy. Increasing the excitation wavelength leads to a decrease in pump photon energy. Consequently, when photon energy is weaker, a greater number of photons is required to ionize an ion, as shown by the increasing value of $\langle x + 1\rangle$. However, the increase in $\langle x + 1\rangle$ implies that greater number for pump photons is needed to ionize one ion, ultimately leading to a reduced number of ionized ions within the medium. This decrease in ionization consequently lowers the free electron density as the excitation wavelength increases [20, 21]. Besides, the plasma defocusing behavior observed in [11] for longer wavelengths, along with the effect of pump light diffraction when using higher wavelengths, also contribute to this phenomenon.

On the other hand, the increase in the plasma electron density corresponding to each $\langle x + 1\rangle$ appears analogous to the peaks observed in ATI [18, 22], wherein electron kinetic energies manifest a distinct peak at the minimum photon energy required for ionization. Generally, as the wavelength of the laser pump increases, the ponderomotive force of the laser field U_P enhances while the photon energy decreases. Consequently, as the wavelength increases, the disparity between U_P and photon energy widens.

For the lowest value of $\langle x + 1\rangle$, this disparity is initially small. In this scenario, U_P contributes to the resonant progression of ATI, this imparts higher kinetic energies to ions, extending ionization and augmenting free electron density. However, as the disparity between the ponderomotive force U_P and photon energy increases, the U_P becomes much higher and then, its role transitions into a predominant pushing effect, resulting in tunneling ionization as can be seen in Fig. 2, *a*, the peaks start to become less pronounced after 1.2 μm for higher $\langle x + 1\rangle$ values where the influence of ATI diminishes. Notably, multiphoton ionization does not impact our scenario since the ponderomotive force U_P, consistently surpasses the photon energy within the studied wavelength range.

The enhancement in plasma electron density could significantly enhance the efficiency of THz generation, since in [13], it was found that the relationship between the power of THz generation and the wavelength of the excitation is determined by two main factors, primarily the plasma electron density and the energy of electron oscillation, which increases in proportion to λ^2.

Fig. 3 displays the calculated plasma electron density values derived from Eq. (4) under the assumption of complete ionization via tunneling effect accompanied by a miniature representation of the logarithmic scale in the upper right corner to better illustrate the decrease in values.

It is evident that the plasma electron density exhibits a continuous decrease consistent with findings from previous studies [11–13]. The values obtained in Fig. 3 using Eq. (4) surpass those acquired through Eq. (3), since the current pump parameters inadequately achieve tunneling ionization required for Eq. (4). However, we opted to utilize Eq. (4) to demonstrate the overarching trend of decreasing plasma electron density under the influence of tunneling ionization.

Overall, the analysis of the presented pump parameters using the Keldysh model reveals an intriguing phenomenon that has not been observed in other works: in the context of ATI, the plasma electron density for specific wavelength ranges increases with the excitation wavelength. These ranges are delineated by the minimum number of absorbed photons required for ionization of the ion $\langle x+1 \rangle$. This contrasts with the typical trend observed in tunneling ionization where plasma electron density generally decreases. Consequently, when dealing with wavelengths in the NIR, it becomes essential to account for both tunneling and ATI processes.

To the best of our current knowledge, the phenomenon of increased plasma electron density for specific ranges of excitation wavelengths has not been discovered theoretically prior to this study. However, experimental results presented in [17] demonstrate this effect. In [17], spectra of noble gas atoms were analyzed under ATI in the Mid-Infrared wavelength range. Excitation wavelengths were varied at λ = 0.8, 1.25, 1.5, and 2 μm. It was observed that the photoelectron count increased with longer excitation wavelengths. This observation supports the hypothesis that ATI plays a significant role in the observed increase in plasma electron density as excitation wavelength increases.

Furthermore, in [11], despite adjusting pump parameters for each excitation wavelength, resulting in decreasing pump intensity as wavelength increased within the range of 1.2 to 2.3 μm, measured plasma electron density (Fig. 1, *a* in [11]) exhibited an increase in specific wavelength ranges: (1.2–1.3 μm), (1.5–1.8 μm), and (2.1–2.2 μm). This

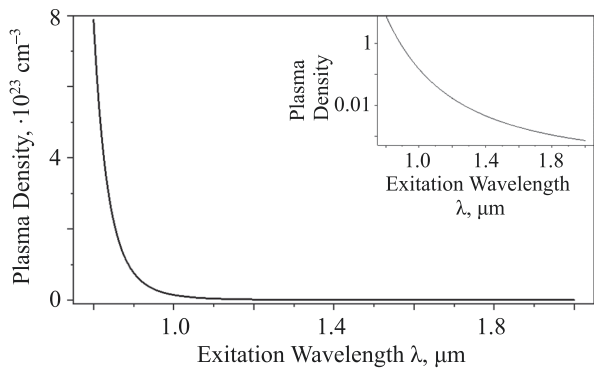

Fig. 3. Plasma electron density ρ_e calculated by integrating Eq. (4) over time as function of the excitation wavelength at a fixed pulse energy of 300 μJ, with a miniature representation of the logarithmic scale in the upper right corner

increase in plasma electron density would become more pronounced at fixed pump intensity. These experimental findings align with theoretical predictions derived from the Keldysh model utilized in this study.

Conclusion

In conclusion, a theoretical study has been conducted utilizing Keldysh theory to analyze the relationship between plasma electron density and the increasing excitation wavelength in dense media. Unlike the typical trends observed in tunneling ionization, the general formula of Keldysh model demonstrates that, under specific parameters, where γ is close to 1, both ATI and tunneling effects can influence the ionization process. We have explained the observed trend and inferred that theoretically, the plasma electron density can increase with wavelength increments within specific wavelength ranges. Moreover, the study offers insights into how to adjust plasma electron density for practical applications by selecting specific excitation wavelengths tailored to the desired plasma electron density.

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