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Varying pre-plasma properties to boost terahertz wave generation in liquids

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Laser-driven nonlinear phenomena can both reveal the structural features of materials and become the basis for the development of various translated technologies, including highly intense terahertz sources. Here we realize a modified single-color double-pulse excitation scheme for enhancing the terahertz wave generation in flat liquid jets, and we show that the pre-ionization effect is crucial for finding the optimal input conditions. The experimental results, being supported by numerical simulations, reveal the preference for longer pre-pulses to induce the effective ionization process and shorter signals for the strong laser-plasma interaction. In addition to the identified features of the terahertz wave energy enhancement with respect to the duration change for both pulses and their ratio variation, we state the possibility of achieving the optical-to-THz conversion efficiency value up to 0.1% in the case of double-pulse excitation of an α -pinene jet.

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Oscillating with a fundamental period of about 1 ps, terahertz frequency radiation can be a perfect tool for the investigation of protein vibrational modes, small molecule rotations, solid-state and gaseous plasma. For over a couple of decades, this electromagnetic range has been considered universal and sufficiently safe for medical applications, products, and environmental ecology quality control¹. Despite the rapid development of terahertz science, both from the fundamental and applied aspects, the problem of developing a highly efficient source of terahertz radiation cannot be considered completely resolved.

As the most relevant approaches to the generation of terahertz radiation, it is fair to highlight those which are based on optical rectification (OR) in crystals², free-electron acceleration (FEL)³, and laser-driven plasmas. All methods have their advantages and limitations, such as, for instance, low damage threshold or the relativistic input energy requirement. The approach based on laser-matter interactions (gas, plasma, solid-state, or liquid) has gained popularity due to its relatively simple experimental implementation, rather a high conversion efficiency values, and wide spectral coverage of the output terahertz field. Further work has focused on the improvement and expansion of this approach. One of the first attempts was to use an external electrostatic field to increase the energy of the generated terahertz waves⁴. Further breakthrough and future work of many scientific groups were associated with the implementation of the so-called two-color scheme, where both fundamental and second harmonics are used as a pump. Two-color filamentation in gases allows obtaining the optical-to-THz conversion efficiency of the order of 0.01%⁵.

The main directions of the laser-driven plasma terahertz source development are the search for an optimal generation medium⁶ (with a high damage threshold, weak terahertz wave absorption, pronounced nonlinear effects) and input conditions variation, e.g., changing pump radiation parameters or modifying the experimental scheme configuration. The former includes the research on the generation of terahertz waves in a wide variety of gases⁷, metals⁸, as well as relatively new works on liquid media^{9–11}. As an example, for input conditions variation, it was shown that the terahertz wave pulse energy could be more than 5-fold enhanced by applying the abruptly autofocusing beam instead of the usual Gaussian one under the same conditions¹². Another promising approach is the pump radiation wavelength shift towards the mid-infrared (IR) range. Thus, the authors of ref. ¹³ demonstrated the possibility of obtaining an optical-to-THz conversion efficiency value up to 2.36% during the two-color filamentation of femtosecond laser pulses at 3.9 μm . Such an impressive value was explained through the stronger photo-currents, longer plasma channels, and additional field symmetry breaking by the generated high harmonics.

In the present work, we investigate the modification of the experimental scheme due to the introduction of double-pulse liquid jet irradiation and bring out the features that distinguish this approach from the single-pulse configuration. Although the double-pump technique itself is not a recent development^{14,15}, the induced pre-plasma effect that manipulates the output energy of the generated terahertz radiation is of considerable interest, motivating further laboratory and theoretical experiments with various input conditions. Thereby, in this study we provide a comprehensive analysis, considering both polar and nonpolar liquid media together with varying pre-pulse and signal pulse duration with their mutual ratio. All this allowed us to achieve the maximum optical-to-THz conversion efficiency value up to 0.1%.

Results

In this work, we use an experimental scheme with a double-pulse laser excitation of a flat liquid jet. Femtosecond p-polarized laser

radiation with a central wavelength of 800 nm and a 1 kHz pulse repetition rate is divided by a beam splitter (BS1) into the reference and signal beams. In contrast to work¹⁴, where a Michelson interferometer was used, in this research a scheme with a Mach–Zehnder interferometer is presented (Fig. 1). This modification is introduced to study the effect of various pulse durations of the reference and signal beam. Their durations are changed by using a dispersion medium of 2–10 cm thicknesses (fused silica). Using the same dispersion medium (QP1, QP2) in both interferometer arms, we avoid the discrepancy in the reference and signal pulse energy characteristics. Thereby, the duration is varied by dispersion broadening and is controlled (measured) by a second-order autocorrelator. The energy of the reference and signal pulses is 0.45 mJ, and their durations are varied from 60 to 350 fs. One of the interferometer arms controls the time delay between pulses from 0 to 30 ps.

A sequence of two collinear pulses falls on a parabolic mirror (PM) with a focal length of 5 cm. The pre-pulse (reference) and signal are focused on a flat liquid jet (see “Methods” section for details regarding liquid jet preparation) at the optimal angle to obtain the most efficient optical-to-THz conversion¹⁶. The estimated laser spot size (FWHM) is 124 μm (the laser intensity is then $2.5 \cdot 10^{13} \text{ W/cm}^2$ for 150 fs pulse). A filter of black Teflon (F) is used to remove visible and IR radiation. Terahertz radiation is then collimated by a TPX lens (L). We register terahertz waves with a standard electro-optical scheme (EOS) based on a 1 mm ZnTe crystal, which allows detecting a signal up to 3 THz.

The inset of Fig. 1 demonstrates the temporal and spectral structures of the generated terahertz field for a single-color double-pulse water jet optical excitation. In this case, the temporal delay is 2 ps, and each pulse duration is equal to 150 fs. The increase in the terahertz field signal can be clearly seen. The “Methods” section provides more details on terahertz radiation energy estimation and pulses separation.

Laser pulse duration effect on the terahertz energy enhancement. Firstly, we compare the optimal value for the pump pulse duration in the case of single- and double-pulse excitation. We conduct experiments to study the terahertz field enhancement considering 2 ps temporal delay¹⁴ and varying pre-pulse and signal pulse durations from 60 to 250 fs (Fig. 2). As a result, in contrast to the pump pulse duration maximum of about 200 fs obtained for the case of single-pulse excitation (Fig. 2a), using a double-pulse scheme, the optimal value shifts to 100–150 fs (Fig. 2b). Moreover, in this case, we obtain a 14-fold enhancement compared to a 4-fold one from the previous results¹⁴.

It turns out that the first observation is the advantage of shorter pulses in double-pulse excitation experiments. The presence of an optimal duration value in single-pulse experiments was earlier interpreted through the combined effect of the electron density exponential growth due to the cascade ionization and pulse energy attenuation with the increase in pulse duration¹¹. The double-pump case is indeed different; it seems reasonable to assume that the optimal pulse duration will experience a shift towards lower values since the important moment is not the effective ionization of the molecules, but rather the strong interaction with a pre-ionized medium.

Optimal liquid for a double-pulse excitation scheme. Since the optimal value for the pump pulse duration was revealed in the previous section, here we establish which of the studied liquids is the most optimal for a double-pulse excitation scheme. Figure 3 shows the results of the signal pulse enhancement in different liquid media excited by pre-pulse and signal of 150 fs, normalized

Fig. 1 The double-pump experimental scheme based on the Mach-Zehnder interferometer. Using the Mach-Zehnder interferometer it is possible to vary the duration of the reference and signal pulses in its arms. This variation is produced by passing the interfering pulses through the quartz plates QP1, QP2 of equal, or different thicknesses (from 2 to 10 cm). An optical delay line is introduced in one of the interferometer arms to control the time delay between the pulses. A sequence of two collinear pulses is then focused by a 5 cm parabolic mirror (PM) on the liquid jet at the optimal angle of incidence. The generated terahertz radiation, collected and collimated by TPX lens L and filtered by a Teflon filter F, is registered on the standard electro-optical scheme EOS. The inset demonstrates the temporal and spectral structures of the generated terahertz field.

Fig. 2 Terahertz radiation energy dependence on the pump pulse duration. The case of single-pulse (a) and double-pulse (b) excitation of a flat liquid jet are compared. Red dots indicate the experimental results (the accuracy of Golay cell measurements is depicted by error bars and is estimated to be 7–10%), black solid lines are for the numerical ones. The shaded region is introduced in order to focus the attention on the pulse duration optimal values obtained in the experiment and numerical simulation.

to the maximum value under the same conditions for water. The curve form, where the preference of a picosecond temporal delay for the liquid media excitation is pronounced, was also revealed by the authors of ref. ¹⁷ and explained by the authors of ref. ¹⁴. Moreover, similar nature of the dependence was obtained in the experimental measurements of X-ray generation in water¹⁸.

In this work, the enhancement range (indicated by the arrows in Fig. 3), presumably corresponding to the plasma lifetime, is revealed to be different for these liquids. In the “Methods” section, we provide additional measurement results on the plasma lifetime by the third-harmonic (TH) enhancement method, which appear to be in good agreement with this conclusion. Thereby,

the double-pump method could shed light on the differences in the molecular response of various liquids in the field of ultrashort IR-pulses. We compare the terahertz pulse energy peak values in the case of each liquid medium and obtain 0.45, 0.37, and 0.33 μJ for α -pinene, ethanol, and water, respectively. Hence, the maximum optical-to-THz conversion efficiency is achieved for an α -pinene jet double-pulse excitation, which is about 0.1%. Similar to previous work, which considered liquid media single-pulse excitation⁹, nonpolar α -pinene demonstrated the highest optical-to-THz conversion due to negligible terahertz absorption and low ionization potential, which leads to the more efficient plasma formation process.

The terahertz wave energy dependence on the pre-pulse/signal pulse duration ratio. Finally, we present the results (Fig. 4) on the pre-pulse and signal pulse duration ratio variation to achieve the terahertz wave maximum energy and thus, define the optimal conditions. The measured terahertz pulse energy duration dependence after the signal beam excited pre-ionized liquid medium is shown in Fig. 4a.

The figure reveals a bright enhancement area around the pre-pulse-to-signal pulse duration ratio of 150:150 fs. Moreover, it demonstrates the advantage of longer pre-pulses to induce the initial electron density and then shorter signal pulses for achieving the maximum energy of a terahertz wave (see the characteristic dashed area in the bottom part of Fig. 4). In order to investigate this result in more detail, we will further study the described dependences using numerical simulation methods and discuss the possible physical mechanisms, which are beyond the experimental results.

Discussion

In this section, we numerically investigate the generation of terahertz radiation upon the excitation of liquid media by two collinear laser pulses to obtain a clearer picture of the pulse duration-dependent features. The equations of strong-field dynamics in the isotropic dielectric medium for a three-band system are used for this purpose¹⁹ (see “Methods” section for some additional details). They can be reduced to one field

Fig. 3 Terahertz wave enhancement dependences on the temporal delay between two collinear 150 fs pulses. The measurements are implemented for water (blue), ethanol (yellow), and α -pinene (red). The accuracy of Golay cell measurements is depicted by error bars and is estimated to be 7–10%. The lines here are introduced as a guide for an eye. The arrows correspond to the temporal enhancement range for each liquid.

Fig. 4 The dependence of the terahertz radiation energy on the pre-pulse/signal pulse duration ratio. **a** Experimentally obtained and extrapolated results. **b** The result of numerical simulations. The dashed line along with white dashed contour (indicating higher terahertz energy area) are introduced in order to highlight the larger pre-pulses—shorter signals preference.

equation, which here is given in its normalized form:

$$\frac{\partial \tilde{E}}{\partial \tilde{z}} - \frac{\partial^3 \tilde{E}}{\partial \tilde{\tau}^3} + \tilde{g} \tilde{E}^2 \frac{\partial \tilde{E}}{\partial \tilde{\tau}} + \tilde{g}_p \int_{-\infty}^{\tilde{\tau}} \tilde{E}^3 \exp\left(-\frac{(\tilde{\tau} - \tilde{\tau}')}{\langle \omega \rangle \tau_c}\right) d\tilde{\tau}' \times \int_{-\infty}^{\tilde{\tau}} \tilde{E}^2 \exp\left(-\frac{(\tilde{\tau}' - \tilde{\tau}'')}{\langle \omega \rangle \tau_p}\right) d\tilde{\tau}'' = 0. \quad (1)$$

Here $\tilde{E} = E/(E_0 \cdot \sqrt{\xi})$, where E is the electric field of radiation, E_0 is the peak amplitude of the spectral-limited pulse at the input surface, and $\xi = \tau_{\text{pulse}}/\tau_{\text{SL}}$ is the ratio of the chirped pulse to a spectral-limited pulse duration to consider the change in a peak amplitude with varying pulse duration; $\tilde{z} = za \langle \omega \rangle^3$, z is the propagation coordinate, a is normal dispersion coefficient, $\langle \omega \rangle$ is central radiation frequency with wavelength $\lambda_0 = 800$ nm; $\tilde{\tau} = \tau \langle \omega \rangle$, where $\tau = t - zn_0/c$ is time in the moving frame of reference, c is the speed of light in vacuum, n_0 is a linear refractive index. Coefficient $\tilde{g} = gE_0^2/(a\langle \omega \rangle^2)$, where $g = 2n_2/c$ and n_2 is the nonlinear refractive index, characterizes low-inertia cubic nonlinear response of the medium, $\tilde{g}_p = g_p E_0^4/(a\langle \omega \rangle^3)$, where $g_p = \frac{2\pi}{c n_0} \alpha \beta$, is introduced to describe the plasma nonlinearity, where $\alpha \beta$ characterizes the efficiency of the electrons transition to the quasi-free states. τ_c and τ_p are the times of collision relaxation and relaxation of highly excited states. All medium parameters are taken according to ref. ¹⁴.

To match the experimental conditions, the normalized radiation field, yielded by two collinear chirped Gaussian pulses with the same energy of 0.45 mJ, a pulse delay of 3 ps and variable pulse duration in the range of 50–300 fs, is used:

$$\tilde{E}(\tilde{\tau}) = \exp\left(-\left(\frac{\tilde{\tau}}{\tilde{\tau}_{\text{pulse1}}}\right)^2\right) \sin(\tilde{\tau} + A_1 \tilde{\tau}^2) + \exp\left(-\left(\frac{\tilde{\tau} - \Delta \tilde{\tau}}{\tilde{\tau}_{\text{pulse2}}}\right)^2\right) \times \sin((\tilde{\tau} - \Delta \tilde{\tau}) + A_2 (\tilde{\tau} - \Delta \tilde{\tau})^2), \quad (2)$$

where $\tilde{\tau}_{\text{pulse1,2}} = \tau_{\text{pulse1,2}} \langle \omega \rangle$ are the normalized durations for reference and signal pulses, respectively; $A_{1,2}$ represents a frequency modulation and is chosen such that the width of the chirped pulse spectrum fit the width of the spectrum for the 35 fs spectral-limited pulse; $\Delta \tilde{\tau} = \Delta \tau \langle \omega \rangle$ is a normalized temporal delay between the pulses.

To begin with, we demonstrate that the results of numerical simulation are in fairly good agreement with experimental studies of the terahertz wave generation during single- and double-pulse excitation of a liquid medium (see Fig. 2, solid lines). The numerical simulation of the terahertz energy enhancement during the liquid medium irradiation with a signal pulse, depending on the mutual ratio of the pre-pulse and signal pulse durations is not

Fig. 5 Terahertz waves generation via double-pulse α -pinene excitation vs. two-color filamentation in air. The comparison of signal (a) and spectral (b) power of terahertz radiation generated during two-color air filamentation and double-pulse liquid jet excitation. The double-pulse technique is implemented for α -pinene liquid for the optimal 150 fs duration (magenta), while in the case of two-color air filamentation we use 35 fs pulse (blue). The optimal parameters are determined for the β -BBO crystal position when a maximum terahertz signal was observed.

that clear (Fig. 4b). The maximum of terahertz radiation energy is obtained for 130–130 fs and 150–100 fs cases with a little dominance of the first one. However, it demonstrates the same trend of long pre-pulse/short signal preference.

Since the generation efficiency of terahertz radiation is varied by the photocurrent, which in turn depends on the induced electron density, we propose an analysis of the terahertz wave energy enhancement during the double-pulse liquid jet excitation mechanism.

Both plasma and Kerr nonlinearities contribute to the terahertz emission. However, with our input conditions, the terahertz emission is dominated by the effects of induced plasma. The research on the comparison of plasma and Kerr nonlinearity contributions in the single-pulse excitation case was already done in ref. ¹⁹. Numerical simulation based solely on considering the third-order Kerr nonlinearity leads to the negligible terahertz generation, while the plasma effect seems to determine this process significantly. Thereby, the first stage in the case of double-pulse excitation can be represented as a microplasma formation and terahertz generation due to the photocurrent and bound electron nonlinearity. This is followed by the terahertz field enhancement stage due to the laser-plasma interaction. Generation of a strong photocurrent is more critical at this stage; thereby, pulses of shorter duration with a higher peak power are preferable. This is consistent with the time evolution of the current density $\frac{\partial j}{\partial t} + \frac{j}{\tau_c} = \beta \rho E^3$. The current is proportional to E^3 and to the number of quasi-free electrons ρ ; consequently, the dependence on the peak intensity is stronger.

The theoretical model based on the density matrix formalism (Eq. (3)) considers no possible reflection of the pump radiation. Thereby, a short signal pulse will only enhance the shielding effect²⁰ and reduce the energy contribution of the pump radiation. This may be a reason for the discrepancy between the experimental and theoretical results in Fig. 4a, b. Moreover, here we can also talk about the possibility of energy redistribution since in the experiment, generation occurs in water and additional nonlinear absorption also happens. Decreasing the pulse duration, we increase the nonlinear absorption, reducing the energy contribution directly in the terahertz generation process.

In conclusion, we propose an effective modified double-pulse excitation scheme with a Mach-Zehnder interferometer to investigate the pre-plasma effect on the terahertz wave energy enhancement in flat liquid jets, and thereby, achieve new conversion efficiency values. Both experimental and theoretical

results reveal that longer pre-pulses are preferable to induce the effective ionization process, while shorter signals—for the strong laser-plasma interaction. This is in agreement with the presence of a pump pulse duration optimal value for the single-pulse excitation case, which origin is determined by the simultaneous cascade ionization processes enhancement and the peak intensity attenuation with the increase in the pump pulse duration. Furthermore, we provide plasma lifetime measurements, which are consistent with THz enhancement traces. In this work, the possibility of achieving the optical-to-THz conversion efficiency value up to 0.1% in the case of double-pulse excitation of an α -pinene jet is demonstrated. It is important to emphasize, that this result, obtained for an 800 nm laser pump, can be scaled further when shifting the central wavelength to a mid-IR region¹³ and reach even more impressive values of about 1–5%. Thus, revealing the optimal conditions for the highly efficient terahertz wave generation in liquids, our study brings the development of powerful and economical terahertz sources closer to reality.

Methods

Liquid jets creation. As a source of a flat liquid jet, we use a system that allows pumping liquid under pressure at 1 m/s speed and forms a plane-parallel flow through the nozzle²¹. In all experiments, a liquid jet with a thickness of 100 μ m is studied. As a generation medium, we choose polar water and ethanol, and nonpolar α -pinene to investigate the effect of the liquid media properties on the optical-to-THz conversion efficiency, which was already essential in the single-pulse case⁹.

Terahertz energy estimation and pulses separation. To estimate the terahertz energy and optical-to-THz conversion efficiency, we use two methods of registration, both EO, and the Golay cell. The latter becomes necessary since it is impossible to separate the pulses with a temporal delay of $\Delta\tau < 2$ ps using EO detection. To obtain the output energy, we integrate the square of the terahertz field amplitude. The conversion efficiency is evaluated when the radiation reaches the detector. Therefore, we consider that after passing the Teflon filter and focusing system, the radiation losses of 10% occur.

To obtain an additional estimate of the optical-to-THz conversion efficiency, we further compare the temporal and spectral structures of the terahertz field generated by α -pinene double-pulse excitation and by the common two-color air filamentation technique (Fig. 5a, b).

The experimental scheme is not significantly modified in this case. For comparison, the duration of the pump pulse is taken to be optimal for the case of two-color filamentation in air and is equal to 35 fs. The filament is generated by focusing the femtosecond pump radiation with a 5 cm lens. The β -BBO crystal with 300 μ m thickness placed behind the lens focus is used for the second-harmonic generation (with 10% conversion efficiency). In our case, we determine the optimal parameters for the β -BBO crystal position when a maximum terahertz signal was observed. In a result, comparing the integrated spectral power for both methods,

Fig. 6 Plasma lifetime measurements via third-harmonic enhancement method. **a** TH emission spectrum in the case of crossing the filaments (blue line) and from the unperturbed filament in water (dashed black line). **b** TH enhancement measurement traces as a function of delay in water (blue) and ethanol (magenta). The traces are normalized to the signal's maximum.

we obtain more than 10-fold enhancement in the case of α -pinene jet double-pulse excitation.

Plasma lifetime measurements. To measure the plasma lifetime in liquids, we apply the third-harmonic (TH) enhancement method, which is proposed and fully described in ref. ²². We use an experimental setup analogous to the one from this work. Two filaments are formed by focusing the pump and the probe beam by a 5 and 10 cm lens and crossed perpendicularly to each other. The probe beam is collimated by a quartz lens with a focal length of 10 cm. The pulse energy in each channel is 0.9 mJ, and the pulse duration is about 40 fs. The pump beam is moved by a motorized delay line with a step of 1 μ m. The spectrum is measured on ASP 100 (Avesta) spectrometer with 1 μ m resolution in the range from 190 to 1100 nm. We use ultraviolet (UV) filters UG-1, UG-5 (Standa), which allow us to obtain the spectral ranges of 240–420 nm, 250–420 nm, respectively. The liquid jet is placed under the angle of 45° relative to the pump and probe beam. The scanning is implemented 140 times for each liquid, after which the data is averaged.

We measure the plasma lifetime for water and ethanol. Using α -pinene jet we get strong evaporation with further damaging of optical elements. These results in a low signal-to-noise ratio. The typical TH spectrum along with the spectrum of enhanced TH is demonstrated in Fig. 6a.

The change in the intensity of TH emission ($\lambda_{TH} = 265$ nm) from water and ethanol depending on the temporal delay is shown in Fig. 6b. Plasma lifetime, which is determined as decay from the peak value to the $1/e$ level, indeed coincides with the terahertz energy enhancement range demonstrated in Fig. 3.

Theoretical model features and approximations. To numerically investigate the generation of terahertz radiation upon double-pulse excitation, we consider the equations of strong-field dynamics in the isotropic dielectric medium for the three-band system¹⁹:

$$\begin{cases} \frac{\partial E}{\partial z} - a \frac{\partial^3 E}{\partial t^3} + g E^2 \frac{\partial E}{\partial t} + \frac{2\pi}{c n_0} j = 0 \\ \frac{\partial j}{\partial t} + \frac{j}{\tau_c} = \beta p E^3 \\ \frac{\partial p}{\partial t} + \frac{p}{\tau_p} = \alpha E^2 \end{cases} \quad (3)$$

The first equation of this system describes the dynamics of the radiation field, considering the dispersion of the linear refractive index and non-inertial cubic nonlinearity, as well as the effect of the induced plasma. The second equation characterizes the evolution of quasi-free electrons current density under the influence of a radiation field, while the third one describes the change in the concentration of electrons of excited energy states, the transition to which is allowed from the ground.

When deriving these equations, the radiation is assumed to be linearly polarized. Since the current density at the output is represented as the time derivative of the induced polarization of the medium, formally, the currents that are perpendicular to the axis of radiation propagation are considered.

In addition, since we use thin liquid jets of 100 μ m thickness for all experimental measurements and the focal length of the effective positive lens induced by a thin water flow is $f = n_0 w_0^2 / 4 n_2 I_0 L \sim 17$ mm, we neglect spatial effects influence on the pulse in Eq. (3).

Data availability

The data used in this manuscript are available from the corresponding author upon reasonable request.

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Author contributions

Experiments were designed by A.N.T. and S.E.P., in close collaboration with X.-C.Z. and performed by A.N.T., S.E.P., and A.O.I. Experimental data were analyzed by A.O.I., S.E.P., and A.N.T. and discussed by all authors. Numerical simulations were carried out by E.A.P. in consultation with S.A.K. The manuscript was prepared by E.A.P. and A.N.T. with discussions and input from all authors.

Competing interests

The authors declare no competing interests.

Additional information

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