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A Retrospective Study: Are the Multi-Dips in the THz Spectrum during Laser Filamentation Caused by THz–Plasma Interactions?

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Abstract: During the process of terahertz (THz) wave generation via femtosecond laser filamentation in air, as well as through the mixing of THz waves with externally injected plasma filaments, THz waves engage in interactions with the plasma. A characteristic feature of this interaction is the modulation of the THz radiation spectrum by the plasma, which includes the generation of THz spectral dips. This information is essential for understanding the underlying mechanisms of THz–plasma interactions or for inferring plasma parameters. However, a current debate exists on the number of THz spectral dips observed after the interaction, with different opinions of single versus multiple dips, thus leaving the interaction mechanisms still ambiguous. In this work, we retrospectively analyzed the experimental appearance of multiple dips in the THz spectrum and found that the current observations of such dips are predominantly a result of the water vapor absorption with a low spectral resolution. Additionally, we observed that altering the acquisition width of the temporal THz signal also influenced the dips' number. Hence, in future research, simultaneous attention should be paid to the following two aspects of THz–plasma interactions: (1) It is necessary to ensure a sufficiently wide time-domain window to accurately represent the spectral dip characteristics. (2) The spectral dips should be carefully distinguished from the water absorption lines before being further studied. On the other hand, for the case of a single dip in the THz spectrum, we also put forward a new viewpoint of the resonance between surface plasmon waves and THz waves, which should also be taken into consideration in future studies.

Keywords: femtosecond laser filamentation; THz–plasma interactions; spectral dips; water vapor absorption; resonance

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1. Introduction

The plasma filament generated by focusing a femtosecond laser in air is an important terahertz (THz) source [1–4]. Currently, apart from research on the mechanism of THz wave generation [5,6], the interaction between plasma and THz waves [6–14], due to effects like the spatial confinement of a THz wave inside the plasma region [15–18], has attracted more and more attention in the community. The plasma filaments that interact with a THz wave can either emit the THz wave [6,7,11–14] or be from another focused laser beam [8–10]. Resulting from this interaction, theoretically or experimentally, modulation traces such as spectral peaks [6,11–13], broadening [14], or dips [7–10] can be left in the radiated THz spectrum.

Reports on the spectral enhanced peak date back to 1993 [13], where the observed THz peak frequency shifted with the plasma density. Later, the enhanced peaks in THz spectra were observed in both single- and two-color laser pumping schemes, e.g., by V. A. Andreeva et al. [6] (Figure 1a), Na Li et al. [12], Xiaomin Qu et al. [19] and Jiayu Zhao et al. [11]. Moreover, THz spectral broadening has also been detected in the study of THz–plasma interactions by I. Thiele et al. [14] (Figure 1b). This spectral broadening might be regarded as another form of spectral enhanced peak away from the original peak.

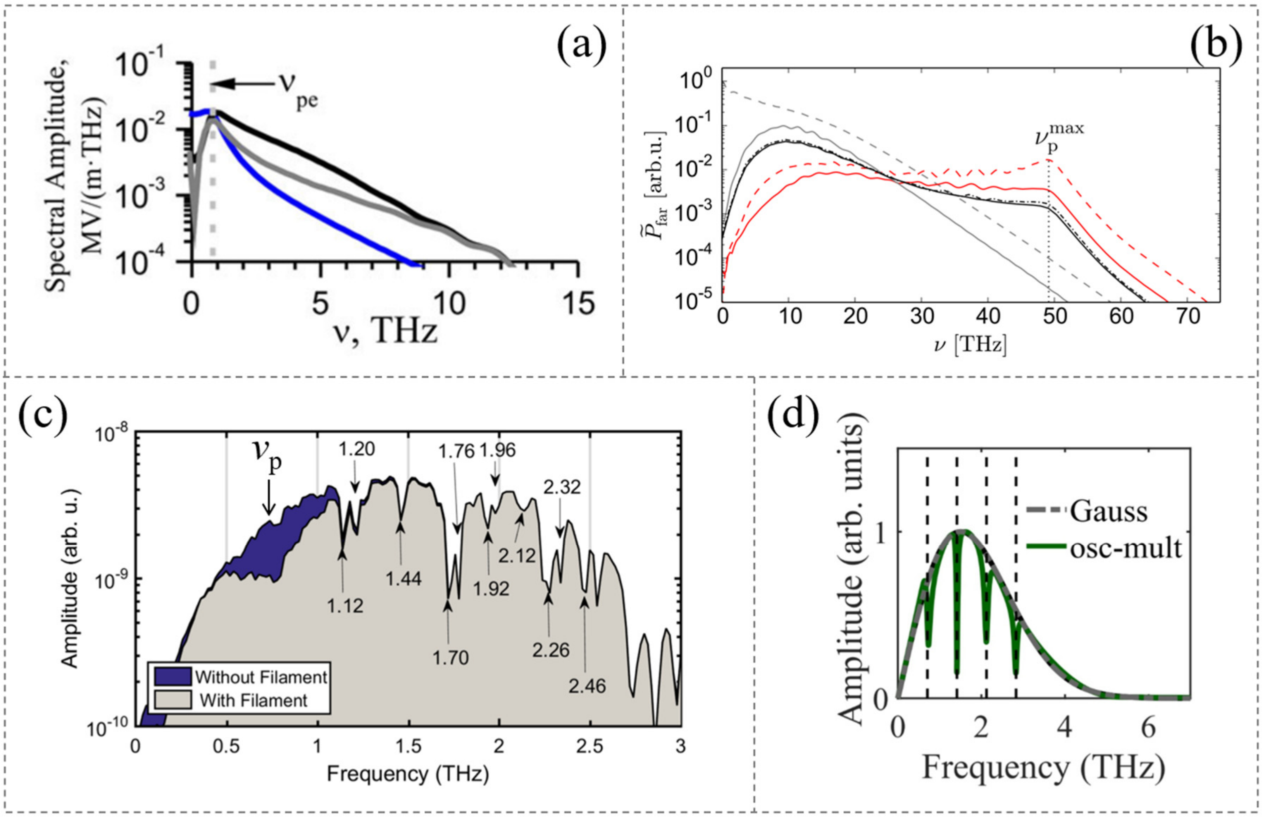

Figure 1. Typical THz spectral profiles post-interaction with plasma filaments. (**a**) An enhanced peak [6], (**b**) a broader width [14], (**c**) a single dip (in blue shadow) [10] and (**d**) multiple dips (the green line) [7]. The reused figure (**a**) adapted from Ref. [6] is used with the permission of the corresponding author and the American Physical Society; (**b**) adapted from Ref. [14] is licensed under CC BY 4.0; (**c**) adapted from Ref. [10] is used with the permission of AIP Publishing; (**d**) adapted from Ref. [7] is used with the permission of the corresponding author and the American Physical Society.

On the other hand, THz spectral phenomena associated with dip features induced by the interaction between plasma and THz radiation have also been frequently reported. Normally, a single dip was produced. For example, Yindong Huang et al. [9] and Zhigang Zheng et al. [10] (Figure 1c) conducted studies characterizing the transmission properties of a THz wave along a laser filament, and demonstrated a significant spectral dip. Furthermore, Xinke Wang et al. [8] investigated the THz spectrum reflected by a pair of cross-filaments and identified a similar singular dip.

The above spectral peak or dip phenomena could coexist, but the displayed main observation depends on the experimental conditions. For example, it is evident that THz spectral peaks and broadening often occur in filamentation-generated THz radiation, while spectral dips frequently appear during the interaction process between THz waves and filaments. Furthermore, the THz spectral dips could also be affected by experimental operations, such as the limited time-domain detection width, which may result in a low-resolution THz spectrum or even the loss of spectral dip information (please see the following sections).

Moreover, these spectral peaks and dips share the same frequency location as the plasma frequency (v_p). Currently, it is widely believed that this characteristic arises from the resonance interaction between plasma and THz waves, i.e., $v_p = v_{THz}$. Considering that the plasma density N_e of laser filaments is in the range of 10^{16-17} cm^{-3}, corresponding to plasma frequencies v_p = 0.9~2.8 THz, it is logical to deduce the feasibility of such resonance effects in the THz frequency band. These THz spectral features resulting from the interaction with the plasma filament are crucial, allowing for not only the inference of physical parameters of plasma (like retrieving N_e by the detected v_p), but also a deeper understanding of the THz–plasma interaction process and underlying mechanisms.

However, very recently, multiple spectral dips due to the plasma–THz interaction (Figure 1d) have also been reported by Nan Li et al. [7]. A theoretical model has been established, inside which the plasma dynamics can result in the formation of an oscillating tail in the THz waveform and dip spikes in the corresponding spectrum. Specifically, the THz radiation is generated from a photoionization net current, which is induced by the symmetry-broken two-color laser electric field. Importantly, these current sources are considered with a Gaussian temporal profile, and their temporal interference has also been calculated. In addition, the current source is assumed to have a phase velocity equal to the laser group velocity, and the plasma oscillation effect is also added in the model. All above considerations can achieve the oscillating tail of the THz radiation in simulations, but the plasma oscillation effect dominates, and this oscillation tail results in multiple dips in the THz spectrum. Experimental evidence has been cited in several references [20–22]. These new results differ from the single dip theory, making the THz–plasma interaction picture unclear. Thus, in this work, we conducted a detailed investigation of the multiple spectral dips in the references [20–22] cited by Ref. [7] in order to reveal their origin.

2. Multiple Dips in the THz Spectrum

According to Ref. [7], THz temporal pulses were extracted from three representative articles [20–22], as shown as (I–III) in Figure 2a. In addition, the signal (IV) is from our work [18]. Note that some of them were temporally shifted or reversed in polarity for being aligned at the peak of the main pulse (along the first vertical dashed line). Impressively, the two valleys of the four signals also coincided at nearly identical temporal coordinates (along the second and third vertical dashed lines).

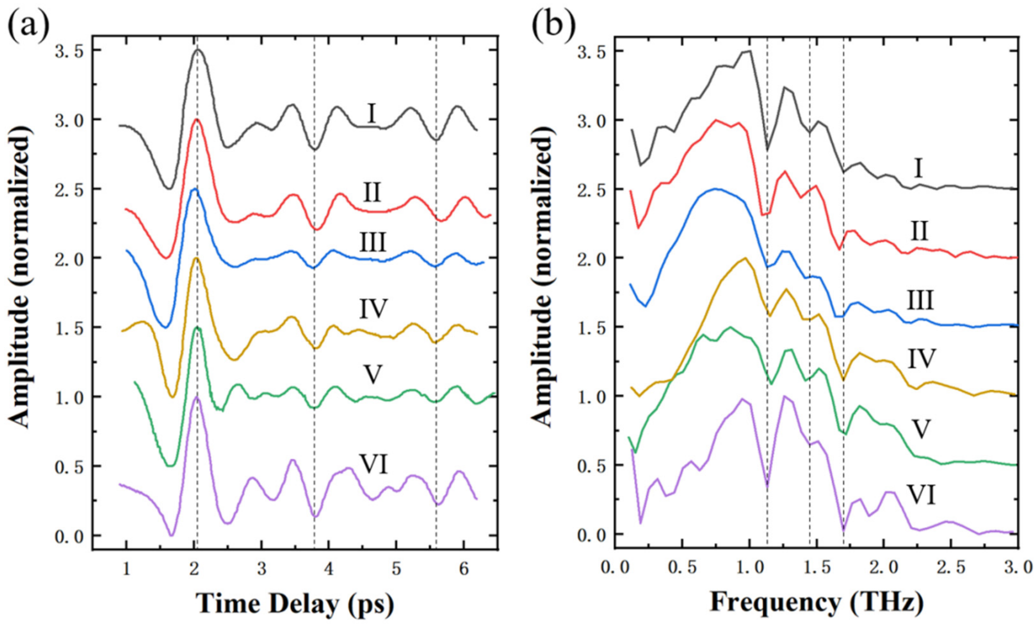

Figure 2. THz signals extracted from the literature [18,20–24] for the (**a**) time and (**b**) frequency domain, respectively. Vertical lines mark the same peaks or valleys of all the signals. Four signals of (I–IV) are from laser-filament-based THz sources, while the other two (V, VI) are from nonlinear-crystal-based THz sources.

These observations further led to similarities between the corresponding four spectra, as depicted as (I–IV) in Figure 2b, which were Fourier-transformed from those in Figure 2a. Three vertical dashed lines are included to emphasize the three spectral dips, which are consistently located at about 1.125 THz, 1.4 THz, and 1.7 THz, respectively.

According to the theory of THz–plasma resonance, these same dips in Figure 2b are indicative of the similar plasma properties of the laser filaments, such as the plasma density. However, these four works had different experimental conditions (as listed in Table 1) for the laser pumping parameters and the plasma filament properties. Therefore, we speculated that these dips may not be related to the interaction between the THz wave and plasma.

Table 1. The experimental conditions for (I–IV) signals in Figure 2 and our work.

Reference	Central Wavelength	Pulse Duration	Single Pulse Energy	Focal Length	Plasma Density	Filament Length
[20]	815 nm	200 fs	25 mJ	150 mm	1.24×10^{16} cm^{-3}	7 mm
[21]	800 nm	80 fs	100 µJ	150 mm	-	-
[22]	800 nm	35 fs	0.65 mJ	100 mm	3×10^{16} cm^{-3}	-
[18]	800 nm	100 fs	1.8 mJ	30 cm	~10^{16} cm^{-3}	2 cm
This work	800 nm	65 fs	5 mJ	100 mm	-	5 mm

For example, we performed experiments under two humidity values of ambient air, with the laser parameters being the same as listed in Table 1, and the THz results are shown in Figure 3. Similar multiple spectral dips can also be seen along the blue dashed lines in the case of large humidity, which seems unrelated to the THz–plasma interactions.

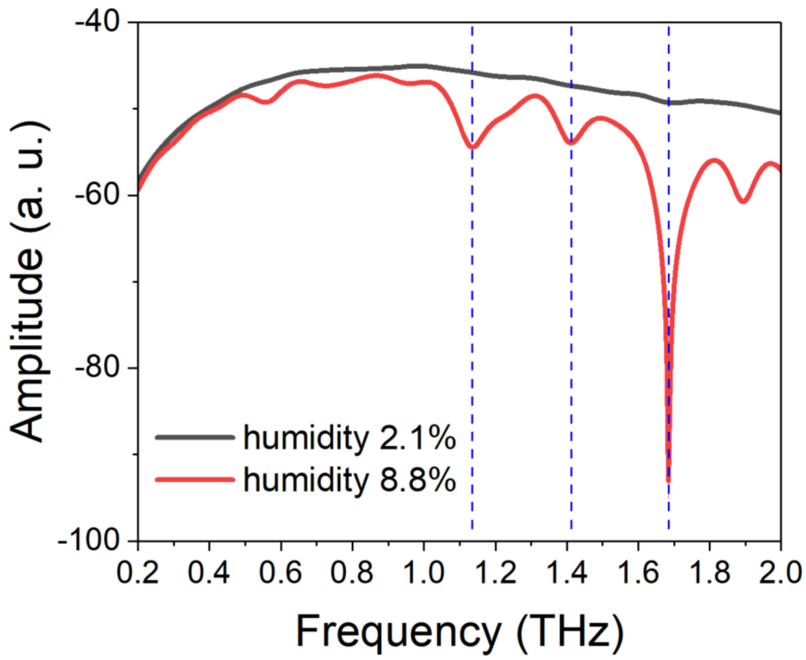

Figure 3. The THz spectra emitted from a two-color laser filament under two humidity values of air.

It is worth mentioning that, although we conducted such an experiment, it cannot cover the extensive range of experimental conditions reported in the literature (as detailed in Table 1). Therefore, retrospectively utilizing and analyzing the data from the literature is more proper in this work, and this is also one of the reasons for these experimental results being made available to the public.

3. Water Vapor Absorption of THz Waves

It is easy to check our speculation above that the spectral dips are not caused by the THz–plasma interactions. Two THz signals from non-plasma sources, namely, lithium niobate [23] and β-BBO crystals [24], are displayed in Figure 2a,b as V and VI, which can be observed in the same manner as the above four signals. This confirms that the multiple dips in the THz spectrum are unrelated to laser plasma filaments. A critical consideration is that the experimental processes of these works all involved the water vapor absorption of the THz waves, which could also create multiple dips in the spectrum. This may account for the observed phenomena in Figure 2.

Therefore, we extracted the water absorption signal from Ref. [21] and performed Fourier transforms on it. The resultant time- and frequency-domain waveforms are shown as (I) in Figures 4a and 4b, respectively. We found that the locations of spectral dips are mainly around 1.12 THz, 1.35 THz and 1.65 THz, which are similar to those in Figure 2b. This strongly suggested a relationship between the spectral dips (Figure 2b) and the water absorption of THz waves. However, the typical water absorption spectrum in Figure 4b exhibits narrow and fine dip structures, while the dips in Figure 2b have much broader widths. Therefore, we subsequently focused on altering the dip width to gain a more comprehensive understanding of the origin of these dips.

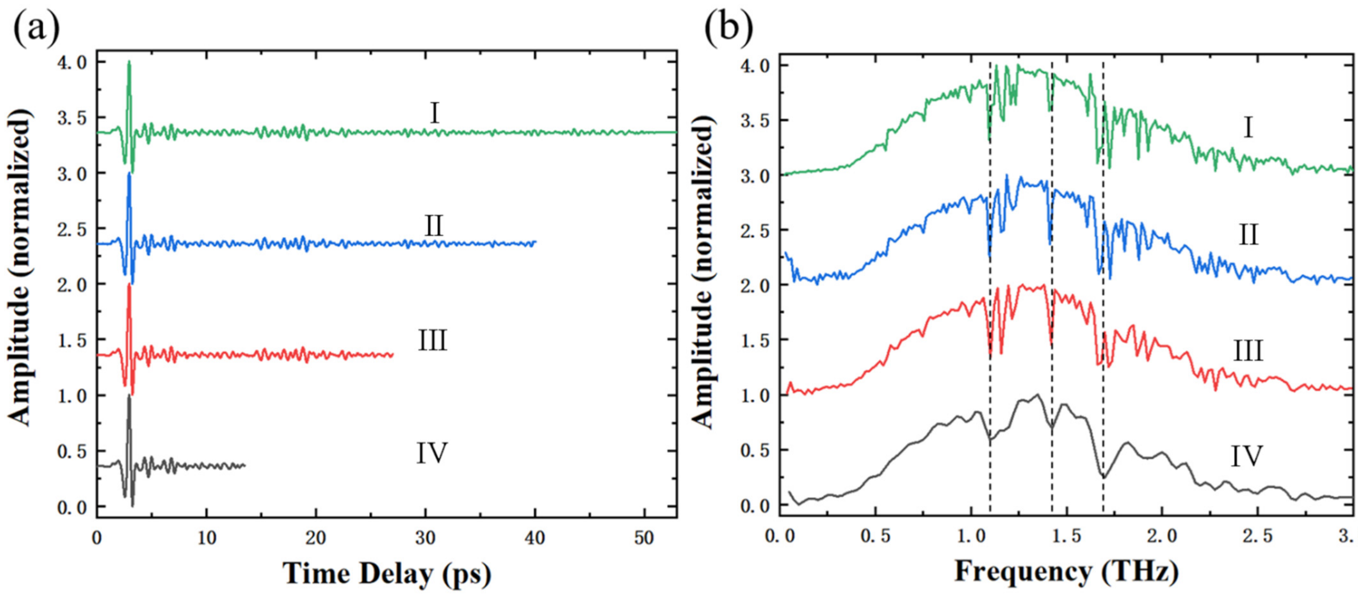

Figure 4. THz signals with the water absorption in the (**a**) time and (**b**) frequency domain, with different temporal widths or spectral resolutions. The vertical lines in (**b**) marked the same spectral dips of all the signals. Data of (I) in (**a**) is from Ref. [21].

We have learned that the spectral resolution is determined by the time-domain window width, which could then change the fine structures of the detected spectra. Thus, we reduced the length of the temporal signal, as shown in Figure 4a (II–IV). Figure 4b shows the corresponding frequency-domain signals. It can be noticed that, as the duration of the time-domain signal was reduced, the dip structures in the water absorption spectra were gradually simplified, which eventually presented broad width features.

Based on these observations, we found that the three dips in the THz spectra extracted from the literature (Figure 2) actually corresponded to water absorption lines in a low-resolution case (Figure 4). This confirms that the experimental multiple dips in THz spectra [20–22] cited by Ref. [7] are not related to the THz–plasma interaction, but are caused by water absorption. One should significantly take the issue of thorough data analyses into consideration when investigating spectral dips that are probably induced by the laser plasma.

4. Artificial Error of Spectral Dips

If the time-domain width of the THz pulse is decreased further, the spectral dips will significantly change, as shown in Figures 5a and 5b, respectively. As for the signal (I), although its temporal width is narrow, the corresponding spectrum still features three dips. However, for the second signal (II) in Figure 5b, two dips (between three peaks) are left. This is attributed to a narrower temporal waveform (II) in Figure 5a, with a main pulse followed by a smaller one.

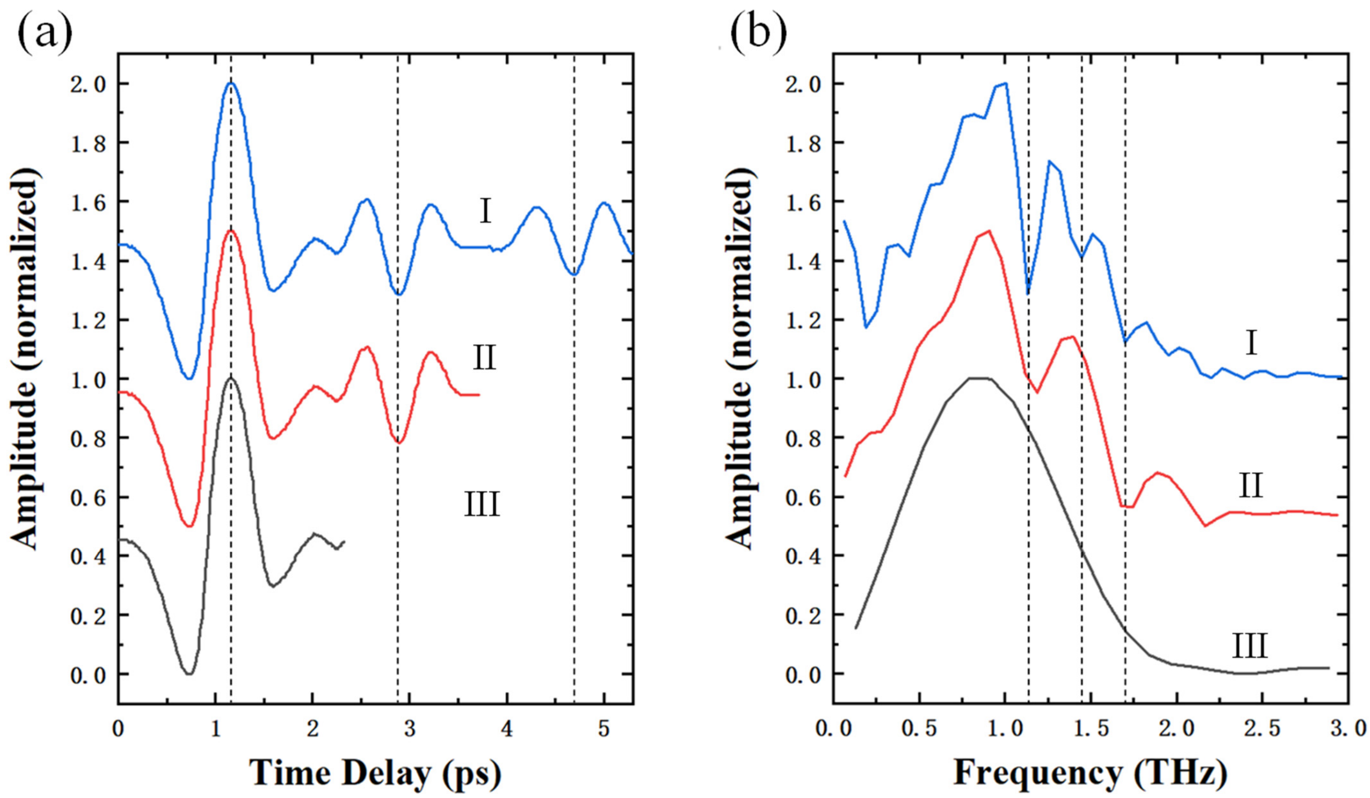

Figure 5. THz signals with water absorption in the (**a**) time and (**b**) frequency domain, with different temporal widths or spectral resolutions. Vertical lines in (**b**) mark the three spectral dips of the top signal. Data of (a-I) is from Ref. [20].

The temporal phenomena (I–II in Figure 5a) do not resemble the traditional water absorption signal, which can lead to potentially incorrect analyses. For example, one might assume this to be caused by the so-called time interference [25] given by the temporal double slits (pulses), or one might mistakenly believe that the second/third pulse is a revival signal given by the nitrogen (N_2), oxygen (O_2) or water (H_2O) molecules in the air [26]. However, in reality, our result is still caused by the common water absorption without any new spectral features, simply because the considered time-domain window width is not long enough. If the THz temporal length is further decreased (into III in Figure 5), no dips can be detected. The above cases can often be found in detection-duration-limited experiments [15], and the THz spectrum with incomplete information is not valid for further analyses.

Based on our analyses, Figures 4 and 5 have revealed two important issues when dealing with THz spectra resulting from THz–plasma interactions, especially with multiple dips: (1) It is necessary to consider whether or not the dips originate from low-resolution water absorption lines; (2) It is essential to ensure the recorded long duration of the temporal THz waveforms after the main pulse in experiments to avoid artificial errors from spectral dips.

5. Resonance between THz and Surface Plasmon Waves

In the above sections, the reported multiple dips in the THz spectra have been proven to result from the water vapor absorption during laser pumping, rather than being induced by the THz–plasma resonance. Therefore, the single dip at v_p is currently the remaining evidence accounting for THz–plasma interactions.

However, considering the spatial scale comparison between laser filaments (tens of micrometers in diameter) and THz waves (hundreds of micrometers in wavelength), it can be expected that the existence of a surface plasmon wave (SPW) in the THz band on the periphery of the laser filament is supported [16,18]. Specifically, as shown in Figure 6, femtosecond laser pulses are focused in the air to create the plasma filament column. Then, the THz wave can be generated within the plasma region, and during its off-axis radiation, it could excite SPW at the plasma–air interface. Figure 6 proposes an internal excitation mechanism of interactions between THz waves and the plasma, which is different from the scheme that the pumping light is usually incident from the external ambient air to the material surface. It is worth noting that Figure 6 does not depict the generation process of THz waves, but rather the interaction of THz waves with the plasma after its generation. During this interaction, the surface-bound THz waves may be created, leading to a spectral defect (i.e., a dip) in the emitted THz spectrum in the air. This represents an attempt to analyze the interaction between THz waves and the plasma filament from the perspective of the surface plasmon.

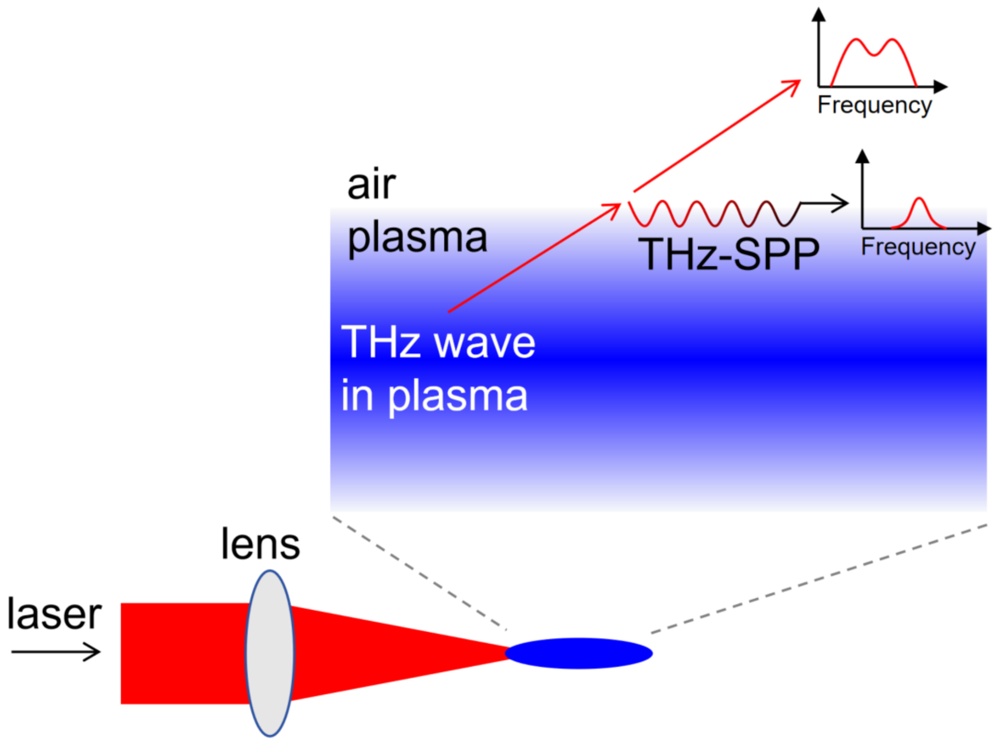

Figure 6. A schematic diagram of THz-SPW generation in the region of a plasma filament column.

The interaction between this surface wave and the THz wave in the plasma also needs to be taken into account. Hence, we next conducted a theoretical analysis of their resonance effect and attempted to derive the resonant frequency position. We adopted the equations of $k_{\mathrm{THz}}^{\mathrm{plasma}} = (w/c)\ (\varepsilon_{\mathrm{THz}}^{\mathrm{plasma}})^{1/2}$ and $k_{\mathrm{THz}}^{\mathrm{spp}} = (w/c)[\varepsilon_{\mathrm{THz}}^{\mathrm{plasma}}/(\varepsilon_{\mathrm{THz}}^{\mathrm{plasma}} + 1)]^{1/2}$, which represent the dispersion relations of the transmitted THz wave and the surface wave in the plasma region, respectively. In both equations, w represents the THz angular frequency; c stands for the speed of light; and $\varepsilon_{\mathrm{THz}}^{\mathrm{plasma}}$ denotes the permittivity of THz waves in plasma, calculated by $\varepsilon_{\mathrm{THz}}^{\mathrm{plasma}} = 1 - {w_p}^2/(w^2 - i\gamma w)$. Here, γ represents the electron collision frequency inside the filament as 0.5–1 THz; w_p is the plasma frequency ($2\pi \times v_p$), calculated by $w_p = (e^2 N_e / m_e \varepsilon_0)^{1/2}$; e represents the charge carried by electrons; N_e represents the plasma density; m_e is the electron mass; and ε_0 is the permittivity of the vacuum.

Subsequently, we calculated the solution for the resonance effect via $\mathrm{Re}[k_{\mathrm{THz}}^{\mathrm{plasma}}] = \mathrm{Re}[k_{\mathrm{THz}}^{\mathrm{spp}}]$, which resulted in $\mathrm{Re}[\varepsilon_{\mathrm{THz}}^{\mathrm{plasma}}] = 0$. From this solution, the relationships of $w^2 + \gamma^2 - {w_p}^2 = 0$ and $w = ({w_p}^2 - \gamma^2)^{1/2}$ are expected. Here, because w_p ($=2\pi \times v_p$) is normally much larger than γ, $w = ({w_p}^2 - \gamma^2)^{1/2} \approx w_p$. Based on this result, we can determine that the resonant frequency between the THz waves and surface waves inside the plasma filament is still $v_{\mathrm{THz}} = v_p$.

Although the resolved resonant frequency (v_p) agrees with the frequently reported value, as mentioned in Section 1, our focus lies in exploring the impact of surface waves on spectral dips, rather than the interaction between plasma and THz waves, as emphasized by previous studies [6–14]. Currently, our theory is not yet fully developed, and this work proposes this idea as a supplementary concept to the existing single-dip theory in the literature. More importantly, it does not change the main focus of our paper: clarifying the source of multiple dips in the THz spectrum. Therefore, we hope to draw more attention by this work to the consideration of the contribution of surface plasma waves to the observed phenomena when analyzing the spectral dips generated by the plasma-based THz sources. We are willing to introduce this new idea in Figure 6 to readers and will carefully validate it in future studies.

It is worth mentioning that there could be overlaps between the multiple dips caused by water absorption and the single dip induced by interactions between THz waves and the plasma. The method to distinguish these phenomena is to eliminate the effect of the ambient water, for example, by purging the experimental setup with dry air. If one wants to further distinguish the two phenomena of the THz wave interacting with the plasma or the surface plasmon wave, one must detect the surface waves. For example, methods like THz-SNOM [27] can be adopted. Furthermore, according to Ref. [28], one can also record the surface wave in free space after it is transmitted along a curved waveguide surface. This work has been planned by us and is underway.

6. Discussion

(1) In this work, we focused on the THz spectrum in order to study the origin of its multiple dips. For this reason, we obtained each THz spectrum via the Fourier transform of a single THz time-domain pulse (from the literature or our work), whose time scale is in the order of picoseconds (ps), much shorter than the plasma lifetime of nanoseconds (ns). Therefore, investigating a single THz pulse or spectrum, and meanwhile taking into account the evolution of the plasma filament, fall beyond the scope of this paper. Actually, during the period of THz pulse generation, the plasma filament can often be considered static without evolution. This approach is frequently adopted in the literature, such as in Ref. [29], which assumes a cylindrical and uniform plasma-density filament for THz emission. On the other hand, we analyzed the THz signals from different plasma filaments under various experimental conditions from different publications, as shown in Table 1 above. These parameters themselves represent a quasi-evolution of the plasma filaments. Therefore, we might consider the studied THz waves in this work as non-static, since they are emitted from many different (dynamic) plasma filaments.

(2) In this work, we did not consider GHz waves to be generations [30–32]. Even if they could interact with the plasma and produce GHz-scale spectral dips, these waves would not appear in the THz spectra we are investigating. This is because the typical spectral resolution of THz experiments is around 20 GHz, corresponding to a time-domain window width of 50 ps. (Frequently, it is more than 20 GHz due to the limited time-domain window width. Here, we do not consider the artificial zero padding at the end of the time-domain signal in order to enhance the spectral resolution after a Fourier transform.) This would result in only five data points within the range of

0–100 GHz (0–0.1 THz) in the THz spectrum. Hence, such a low spectral resolution is insufficient to resolve any GHz dips. In contrast, the THz dips we analyzed are much higher than the GHz range, and can be clearly observed on the THz spectrum (Figure 2).

7. Conclusions

In summary, in this work, we suggested that the theory of multi-dips left in the THz spectrum during THz–plasma interaction still requires experimental verification, as the experimental findings within the existing literature cannot yet confirm this theory. In future studies of multiple dips in THz spectra, in order to understand the THz–plasma interaction, one should first carefully exclude possible water absorption. Following this, it is essential to gather comprehensive THz signal information from extended temporal waveforms to avoid the artificial error of spectral dips. By contrast, currently, the reported single dip effect at v_p given by the THz–plasma resonance remains applicable. Additionally, in this case, we demonstrated that the role of the surface plasmon wave should not be overlooked, since its resonant frequency with the THz wave in plasma is also around v_p.

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Conflicts of Interest: Author Li Lao was employed by the company Tera Aurora Electro-Optics Technology Co., Ltd. The remaining authors declare no conflict of interest.

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