

1 A comprehensive diagnostic of liquid sheet 2 targets for laser ion acceleration

3 Ziyang Peng¹, Zhengxuan Cao¹, Xuan Liu¹, Yinren Shou⁴, Jiarui Zhao¹, Shiyu Chen¹,
4 Ying Gao¹, Pengjie Wang⁵, Zhusong Mei¹, Zhuo Pan¹, Defeng Kong¹, Shirui Xu¹, Zhipeng
5 Liu¹, Yulan Liang¹, Tianqi Xu¹, Tan Song¹, Xun Chen¹, Qingfan Wu¹, Yujia Zhang¹, Zihao
6 Zhang¹, Xueqin Yan^{1,2,3}, Wenjun Ma^{1,2,3*}

7 1 State Key Laboratory of Nuclear Physics and Technology, Key Laboratory of HEDP of
8 the Ministry of Education, CAPT, Peking University, Beijing, China

9 2 Beijing Laser Acceleration Innovation Center, Beijing, China

10 3 Institute of Guangdong Laser Plasma Technology, Guangzhou, China

11 4 Center for Relativistic Laser Science, Institute for Basic Science, Gwangju, Republic
12 of Korea

13 5 Institute of Radiation Physics, Helmholtz-Zentrum DresdenRossendorf, Dresden,
14 Germany

15 Email: pengjiang_123@stu.pku.edu.cn

16 Corresponding author: wenjun.ma@pku.edu.cn

17 Abstract

18 To meet the demands of laser ion acceleration at a high repetition rate, we have
19 developed a comprehensive diagnostic system for real-time and in-situ monitoring of
20 liquid sheet targets (LST) generated by the collision of two liquid jets. The spatially
21 resolved rapid characterizations of a LST's thickness, flatness, tilt angle and position
22 are fulfilled by different subsystems with high accuracy. With the help of the
23 diagnostic system, we reveal the dependence of thickness distribution on collision
24 parameters and the determinants of LSTs' flatness and tilt angle, which are essential
25 for applications of laser-driven ion acceleration and others.

26 **Keywords:** liquid sheet targets; diagnostic; laser ion acceleration

27 Introduction

28 The interaction of ultra-intense laser pulses with micrometer-thin film targets
29 can accelerate ions to very high energy within a few micrometers. Near-100-MeV
30 protons and 1.2-GeV Au ions have been generated by laser acceleration[1][2]. This
This peer-reviewed article has been accepted for publication but not yet copyedited or
typeset, and so may be subject to change during the production process. The article is
considered published and may be cited using its DOI.

This is an Open Access article, distributed under the terms of the Creative Commons
Attribution licence (<https://creativecommons.org/licenses/by/4.0/>), which permits
unrestricted re-use, distribution, and reproduction in any medium, provided the original
work is properly cited.

10.1017/hpl.2023.101

31 novel ion acceleration method attracts widespread attention as it can produce
32 ultrashort ion pulses in a short distance, which is highly appealing for applications
33 such as proton imaging, pulsed neutron generation, and FLASH radiotherapy[3][4].
34 Many of them require a high average flux, which should be achieved by shooting
35 targets at a high repetition rate. However, solid films, the most widely used targets in
36 previous studies, are not suitable for long-term high repetition rate shooting. After
37 each laser-target interaction, the film in an area of about a few mm² around the
38 focal spots would be completely destroyed. If they are shot at kHz, for example,
39 supplying the targets in the vacuum would be a big challenge, not to mention the
40 cost and debris damage to the optical components in the target chamber.

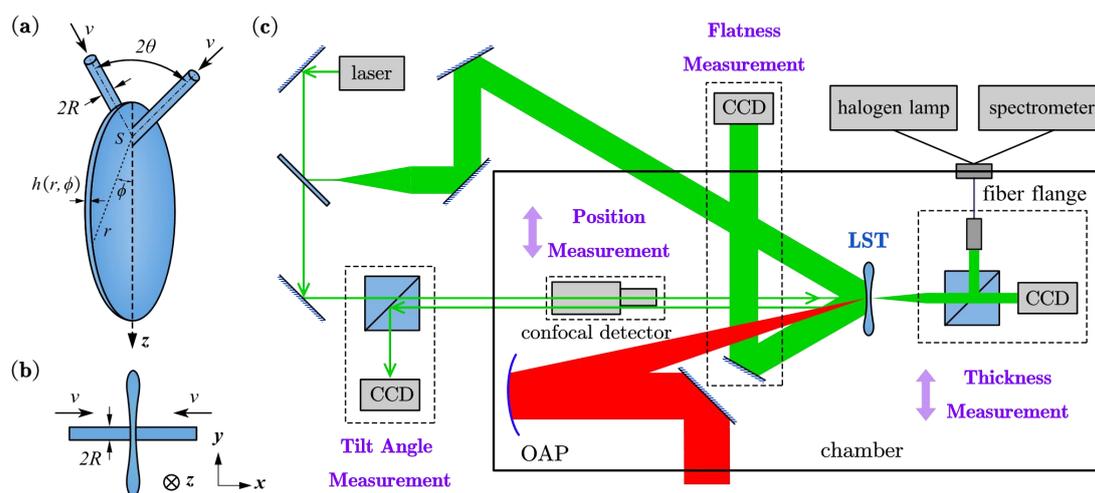
41 Superior to solid films, free-flowing micrometer-thin liquid sheets are promising
42 targets for high-repetition-rate laser ion acceleration[5]. They are made of
43 continuous liquid flow without any supporting substrate. Under the irradiation of an
44 ultra-intense laser pulse, the damaged area can be repaired spontaneously in 0.1 ms,
45 enabling continuous shooting at a repetition rate up to kilohertz or higher[5].
46 Therefore, liquid sheet targets (LST) draw increasing interest in the field of laser ion
47 acceleration.

48 While still in its early stages, some pioneering work had successfully employed
49 LST in experiments. In 2018, Morrison et al. first achieved the generation of 2.3 MeV
50 protons from LST at kHz repetition rate for the first time[6], and a few years later
51 Valdes et al. obtained 3.5-MeV protons from LST at a higher vacuum degree[7].
52 Deuterons with energies up to 4.4 MeV were also generated by using LST made of
53 heavy water[8]. Nevertheless, these works lack a complete characterization of LSTs. It
54 has been known that the film's position with respect to the focal spot and its
55 thickness significantly influences the energy of ions. The tilt angle and flatness of the
56 film would affect the direction and divergence of the ion beam. Therefore, for
57 optimized and stable ion acceleration, comprehensive characterization of films is
58 very necessary.

59 In this work, we present an online diagnostic system for the comprehensive
60 characterization of liquid sheets used in laser-driven ion acceleration. First, an
61 overview of our LST generation and characterization system is presented. Afterwards,
62 the components of the system for the measurement of LST's thickness, flatness, tilt
63 angle and spatial position are described separately in 4 sections. Analysis methods
64 are also given. At last, a summary and future prospects will be provided in the final
65 section.

66 **LST Generation and Characterization System**

67 The liquid sheet was generated with our homemade colliding-jets device, and
 68 relevant information can be found in our previous publication[9]. Fig.1(a) shows the
 69 formation process of a liquid sheet: two liquid jets eject from a capillary tube with a
 70 diameter of 50 μm and collide at a speed of approximately 20 m/s, forming a
 71 collision angle of 60° (2θ). After the collision, the lateral momentum of the two jets
 72 cancels out each other and forms a closed liquid sheet in the orthogonal direction
 73 due to the effect of surface tension, as shown in Fig.1(b). In this work, we have made
 74 improvements to the device, with a total of eight degrees of freedom to regulate the
 75 sheets, including six dimensional translation and rotation, and the relative
 76 two-dimensional adjustment between the two jets. These degrees of freedom are
 77 conducive to stable control and precise characterization of liquid sheets. The liquids
 78 were driven by two high-performance liquid chromatography (HPLC) pumps
 79 (Shimadzu, LC-20ADXR) and injected into two capillary tubes respectively. The back
 80 pressure is usually several tens MPa, depending on the flow rate, capillary length,
 81 and viscosity of the liquid. In this article, the data are obtained by default using a
 82 glycerol aqueous solution with a mass fraction of 50%. Glycerol aqueous solutions
 83 with a mass fraction between 30% and 70% can operate stably in our system, but the
 84 sheet properties may vary. This type of liquid is cheap, non-toxic, and has a low
 85 saturated vapor pressure, making it ideal for LSTs.



86 Fig.1 (a)Schematic of the liquid sheet generation, (b)Top view of the liquid sheet generation,
 87 (c)Overview of the main modules of the diagnostic system.

89 Fig.1(c) is an overview of the diagnostic system, which allows for real-time and
 90 spatially resolved characterization of LST's thickness, flatness, tilt angle and position
 91 fulfilled by different subsystems. The dashed boxes represent these subsystems, and

92 the purple words next to them describe their functions. The solid box represents the
93 vacuum chamber, with LST located near the center of the chamber in fig.1(c). The red
94 laser represents the main laser, which is focused on LST by an OAP. The green light on
95 the left side of LST represents a monochromatic probe light, while the green light on
96 the right side represents a broad-spectrum probe light.

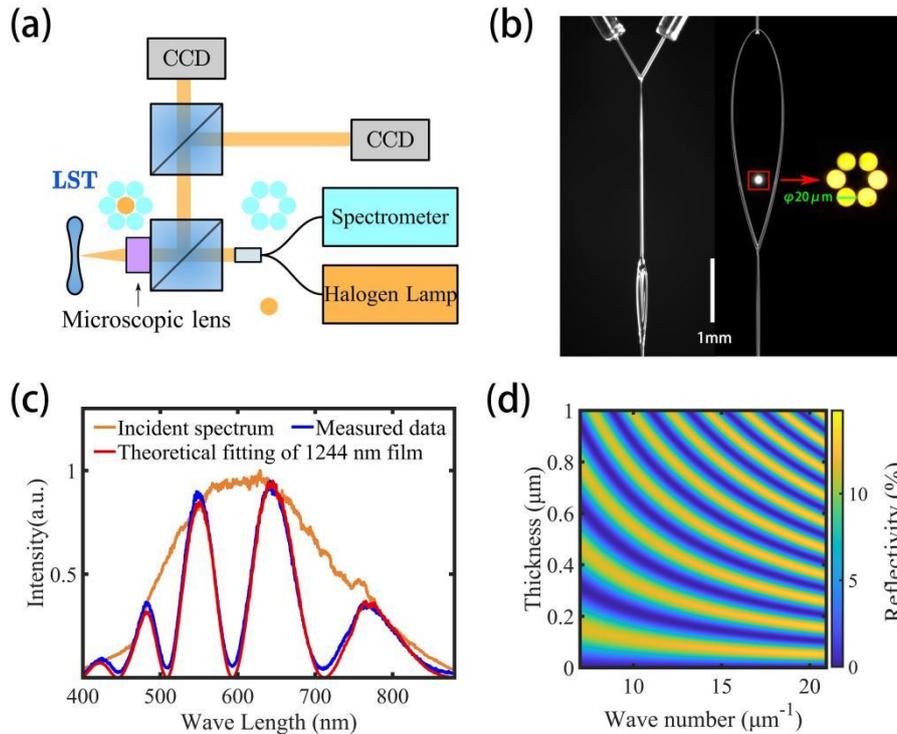
97 It should be noted that fig.1(c) is not drawn to scale. On the left side of LST, only
98 the confocal detector is very close to the sheet (about 20 mm), and it cannot work
99 simultaneously with tilt angle measurement. Typically, the confocal detector is
100 moved away by a motor after the position measurement, so there are actually no
101 obstacles on the optical path of the main laser, and the system can operate for small
102 f-number OAP ($f/1.5$, for example). Further details of the subsystems will be
103 presented in the following sections sequentially.

104 **Thickness Measurement**

105 The thickness of the targets significantly affects ion energy and even the
106 acceleration mechanism[10]. Therefore, it's essential to measure the thickness of the
107 liquid sheet accurately. This could be challenging because LSTs are self-supporting
108 and unable to withstand external force, making contact measurements inapplicable.
109 Besides, the thick edge of the sheet also hinders thickness measurement using side
110 imaging[11]. To address these challenges, we measure the thickness of a LST using
111 the reflected white light spectrum from the film surface. In the following text, we will
112 refer to this method as reflectance spectroscopy.

113 The schematic diagram of our device can be found in the upper right corner of
114 fig.1(c). The halogen lamp and spectrometer are both placed outside the chamber,
115 and two optical fibers are bunched on the feeding flange and connected to the
116 chamber. Fig.2(a) is a more detailed diagram: the white light emitted from a halogen
117 lamp is focused by the microscope lens and then irradiated on LST, and the reflected
118 light is sent back to the spectrometer via optical fibers behind the lens. The
119 microscopic lens enables simultaneous imaging of LST, while white light provides
120 illumination. In order to increase the signal, six optical fibers are used to collect
121 reflected light, they hexagonally surround the central fiber that connects to the
122 halogen lamp. Fig.2(b) shows the side and front view of LST, where the bright dot in
123 the center of the sheet is the incident white light. The light source is connected to
124 the six fibers here to clearly display the structure of the fiber bundles. Ordinarily, the

125 central fiber functions as the light source, while the six outer fibers gather reflected
 126 light signals, as depicted by the six blue dots in fig.2(a). The diameter of each fiber is
 127 200 μm , and the spot focused on the sheet is approximately 20 μm . In the next
 128 section, it will be seen that the thickness of the liquid sheet can be considered
 129 uniform in such a small area.



130
 131 Fig.2 (a)Schematic diagram of reflectance spectroscopy, (b)Side view and front view of LST,
 132 the light source is connected to the six cores here to clearly display the measurement point,
 133 (c)A typical reflection spectrum of LST with the theoretical fitting curve, (d)Theoretical
 134 reflectivity of films with different thicknesses at different wave numbers.

135 Interference between the two surfaces of the film results in a chirped oscillation
 136 in the reflected light spectrum (it displays gradually changing periods, with higher
 137 frequency at short wavelengths and lower frequency at long wavelengths). The

138 reflectivity follows equation (1), where $\delta(h, \lambda, \theta) = \frac{4\pi n(\lambda)h \cos \theta'}{\lambda}$ is the phase

139 difference, λ is the wavelength of light, θ is the angle of incidence and θ' is the
 140 refraction angle in film, $R_0(\lambda, \theta)$ is the interface reflectivity determined by Fresnel
 141 law, h is the thickness of film and $n(\lambda)$ is the refractive index of the material[12]:

$$142 \quad R(h, \lambda, \theta) = \frac{4R_0(\lambda, \theta) \sin^2[\delta(h, \lambda, \theta) / 2]}{[1 - R_0(\lambda, \theta)]^2 + 4R_0(\lambda, \theta) \sin^2[\delta(h, \lambda, \theta) / 2]} \quad (1)$$

143 The blue curve in fig.2(c) shows a typical measured spectra from a LST, which can
 144 be well fitted by assuming a thickness of 1244 nm. However, the fitting method is

145 time-consuming and therefore difficult to use in real-time thickness measurement.
 146 Reflectance spectroscopy reported before mostly used peak positions to calculate
 147 the thickness[13], which was highly efficient, but this resulted in low information
 148 utilization and lack of accuracy.

149 Therefore, we develop a fast algorithm for accurately extracting thickness
 150 information from reflection spectra, and the principle is as follows: Fig.2(d) gives
 151 theoretical reflectivity values at $\theta = 0$ as functions of thickness and wave number.
 152 One can see that for films with thicknesses ranging from hundreds of nanometers to
 153 several micrometers, there are multiple peaks in the spectral range of 300-1000 nm
 154 for each thickness, and the oscillation interval in the wave number space is constant
 155 and proportional to the thickness. This can be proven by equation (1), the reflectivity

156 exhibits a periodic oscillation, with an instantaneous frequency of $\nu(\lambda, h) = \frac{2n(\lambda)h}{\lambda^2}$,

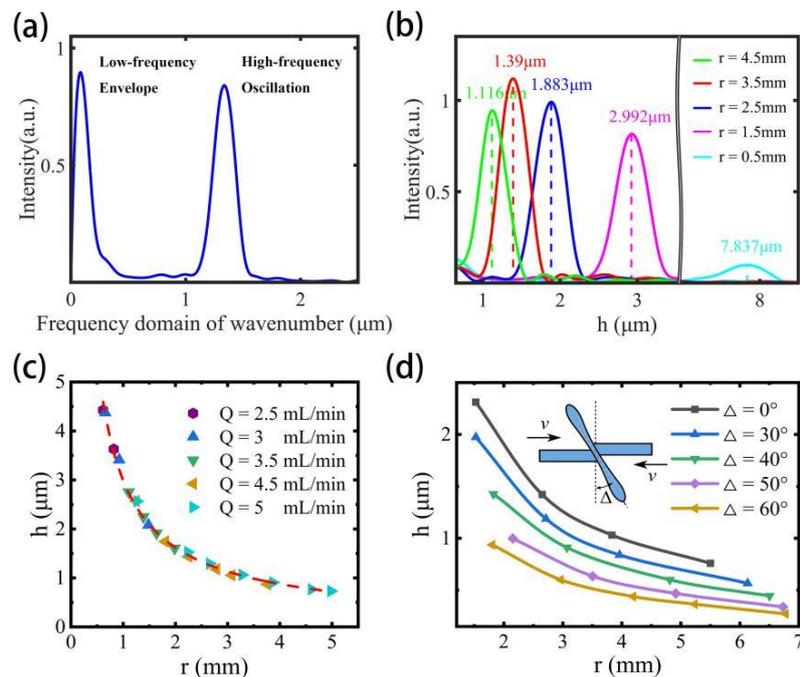
157 and in k-space, it is $\nu(k, h) = \frac{n(k)h}{\pi}$. This can be considered a constant if the film is

158 uniform and the dispersion curve is known. By performing Fourier transform on the
 159 reflection spectrum and its peak directly corresponds to the thickness.

160 In fig.3(a) we show an example of the Fourier transform of the measured
 161 spectrum in wave number space. There are two peaks in the frequency domain, the
 162 low-frequency envelope and the high-frequency oscillation of the spectrum,
 163 respectively. The high-frequency peak position is proportional to the thickness, while
 164 the low-frequency peak is caused by the slowly changing envelope of the spectrum,
 165 as shown in the incident spectrum in fig.2(c). It should be clarified that the incident
 166 spectrum can only be used as a relative reference. We can normalize the measured
 167 spectrum by the incident spectrum to minimize the impact of the low-frequency
 168 envelope, but this does not ensure getting absolute reflectivity. It is precisely because
 169 of this that calculating the film thickness through the peak positions of the reflection
 170 spectrum is inaccurate, as the peak positions are modulated by the spectrum
 171 envelope. The inherent low-frequency component sets a lower limit for the
 172 measurement (when the oscillation frequency is close to the envelope frequency, the
 173 oscillation will be masked by the envelope). In our system, the minimum measurable
 174 thickness is about 200 nm, which is competent for the current LST (the reported
 175 thinnest liquid sheet produced by colliding-jets is 450 nm). The accuracy of this
 176 method can reach ± 5 nm, and in most cases the error does not exceed 2 nm.

177 Based on this method, we measured the thickness of LST at different flow rates
 178 and positions. As defined in fig.1(a), we will use these symbols in the following text:
 179 h means the thickness of LST, r is the distance to the colliding point, and ϕ is
 180 the azimuth angle of the colliding point. In fig.3(b), five spectra measured at different

181 positions on LST (all five measured points here are $\phi = 0$, r takes different values)
 182 were transformed into frequency domain. The abscissa was linearly converted to
 183 thickness, and we call it the thickness spectrum. Note that the intensity of the
 184 thickness spectrum does not have a special effect, only the peak position is helpful.
 185 As the distance from the collision point increases, the liquid sheet gradually becomes
 186 thinner, and the broadening of its thickness spectrum is mainly caused by limited
 187 measurement bandwidth and dispersion, while at a distance closer to the collision
 188 point, it is mainly due to the superposition of multiple thicknesses within the
 189 measurement point (with a large thickness gradient, as will be seen in the next
 190 section).



191
 192 Fig.3 (a)Fourier transform of a measured spectrum in wave number space, (b)Thickness
 193 spectrum at different positions on LST, (c)Thickness of LST at different flow rates and
 194 positions, (d)Thickness of LST under different misalignment of colliding.

195 The overall results are shown in fig.3(c), the thickness under different flow rates
 196 seems to fall on the same curve, which means that the thickness at the same
 197 position is almost not affected by the flow rate (not exact. The measurement at fixed
 198 points indicate that the flow rate will have a slight impact on the thickness). For our
 199 application, we always hope to increase the flow rate as much as possible to
 200 generate a thinner sheet, but this is limited by factors such as system pressure and
 201 fluid instability, and usually cannot exceed 6 mL/min. Another way to obtain a
 202 thinner liquid sheet is colliding the two jets with a misalignment[14][15], as shown in
 203 the diagram in fig.4(d). Different misalignment leads to different deflection angle Δ ,

204 and as the angle increases, the sheet also becomes thinner. We generated a liquid
205 sheet about 240 nm at $\Delta = 60^\circ$, and this is the thinnest liquid sheet formed by two
206 colliding jets to date.

207 In summary, we developed an algorithm and device for online measurement of
208 LSTs' thickness and it's combined with imaging system in a very compact way.
209 Accuracy for most LSTs is typically within ± 2 nm, and for LSTs around 250 nm, the
210 error is ± 5 nm. Additionally, we have a practical method for measuring films below
211 200 nm, and its details will be described in the "Tilt angle measurement" section.

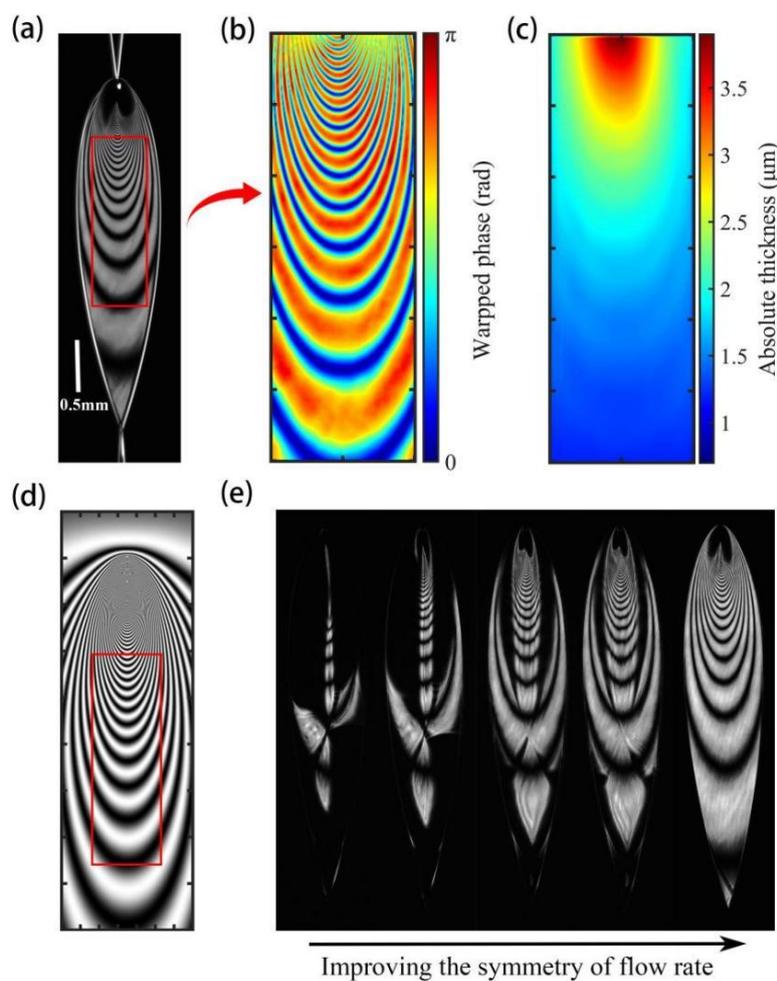
212 Flatness Measurement

213 Reflectance spectroscopy measures the thickness at a specific point, and overall
214 knowledge of the thickness distribution and flatness of a LST is also important. It
215 helps us monitor the quality of the as-produced LST and choose the optimal shooting
216 position. In our diagnostic system, an expanded monochromatic laser (indicated by
217 the thick green path in fig.1(c)) is obliquely illuminating the liquid sheet, and a CCD is
218 set in the reflection direction to image the interference fringes of the sheet, which
219 can be used to obtain the contour information of the LST's thickness [16]-[21].

220 Fig.4(a) shows a typical pattern of interference fringes. In work[16][18], the local
221 one-dimensional thickness variation is analyzed from the interference fringes, but
222 the two-dimensional thickness distribution has not yet been obtained. Theoretically,
223 the sine value of the phase can be derived from interference fringes, but it is not
224 feasible to extract the thickness distribution due to blurriness in the phase. However,
225 after analysis, we have confirmed that reconstructing the thickness distribution is still
226 possible for liquid sheets. Here we present the first recovered two-dimensional
227 thickness distribution of the liquid sheet. First, we assume that the intensity
228 distribution of the expanded beam on such a small sheet is uniform, so the
229 brightness of the interference fringes is solely related to the reflectivity of the LST.
230 For fixed wavelength and incident angle, the wrapped phase is a single-valued
231 function of thickness. Thus, we can uniquely obtain $\sin \delta$ according to equation(1).
232 δ is hidden in $\sin \delta$, so the obtained phase is wrapped in the range of 0 to π , as
233 shown in fig.4(b). Since the thickness along the axis of the liquid sheet (as shown in
234 fig.3(c)(d)) continuously decreases from top to bottom, so the thickness at every
235 interference fringe is $\lambda / 2n$ thicker than that at the adjacent fringe below. Based on
236 the prior information, δ can be unwrapped using some classic algorithms[22][23].
237 Fig.4(c) depicts the retrieved 2D thickness distribution of the LST, where the absolute
238 thickness value is determined through the reflectance spectroscopy measurement at

239 a single point.

240 It can be seen that the thickness in the upper part changes significantly
 241 compared to the relatively flat lower part, where the variation is less than 1 nm
 242 within a few micrometers. The spot of main laser in laser ion acceleration is usually
 243 several micrometers, which means the change in LST's thickness within the laser
 244 focal spot is less than 1%. This property makes the lower part more suitable for laser
 245 ion acceleration, as it provides a flat surface with a very thin thickness. Research has
 246 shown that thin films with a thick middle and thin sides may be more suitable for
 247 ultra-intense laser ion acceleration[24], while LSTs have similar properties and can be
 248 selected at locations with different thickness gradients as needed.



249

250 Fig.4 (a) Typical interference fringes of LST, (b) Wrapped phase in the red box of (a),
 251 (c) Thickness distribution (μm) unwrapped from (b), (d) Interference fringes corresponding to
 252 the thickness distribution of the Hasson's model, (e) The change of interference fringes with
 253 the improvement of flow symmetry.

254 Interference fringes are not visible near the collision point in fig.4(a), and we
 255 used Hasson's model[25] to calculate interference fringes, as shown in fig.4(d). The

256 thickness distribution given by the Hasson's model is as follows:

$$257 \quad h = \frac{R^2 \sin^3 \theta}{r(1 - \cos \phi \cos \theta)^2} \quad (2)$$

258 From the equation and fig.4(d), it can be seen that the theoretical interference
259 fringes closely align with the measured fringes, and the thickness variations are
260 significantly drastic in the vicinity of the collision point. In fig.4(d), due to numerical
261 resolution, there are two blurry shadows corresponding to the invisible area in
262 fig.4(a), as these fringes must be captured at a very precise angle. Based on the
263 numerical aperture and working distance of the lens in our system, when the angle
264 of reflected light from LST deviates by 24.6 mrad, it cannot be collected. When the
265 gradient of thickness variation exceeds 50 nm per micrometer and causes the sheet
266 surface to tilt beyond the collection angle, the fringes can not be captured.

267 This phenomenon not only occurs near the collision point, if the surface of the
268 sheet bends and no longer behaves like a mirror, the integrity of the interference
269 fringes will be damaged. The unevenness of LST can occur due to various factors, and
270 fig.4(e) shows a case caused by asymmetry of flow rate. The sheet is bent and ripples
271 appear on the surface, the thickness gradient at some positions reaching hundreds of
272 nm per micrometer. The asymmetry of the leftmost subgraph reaches 3%, gradually
273 improving to the right until it is completely symmetrical. The asymmetry of the flow
274 rate has such a significant impact on the flatness of LST, so we use two pumps to
275 independently control the two jets, avoiding asymmetry and instability caused by
276 three-way splitting. In fact, we find that different asymmetric factors can cause
277 different bending modes of the liquid sheet, including non-uniformity of mixed
278 solutions, misalignment collision of the jet, asymmetry of flow rate, uneven end face
279 of capillary outlet and so on, which will reflect in the interference fringes. This
280 feature allows real-time monitoring of the flatness of LST as the sheet is always in a
281 changing state. This significantly contributes to the stability and controllability of
282 laser ion acceleration.

283 In summary, we realized the reconstruction of the two-dimensional thickness
284 distribution of LST through the interference fringes for the first time, and the
285 thickness of the flat area varies monotonically with a gradient less than 0.5 nm per
286 micrometer. Based on the pattern of the fringes, diagnosis and feedback of stable
287 operation was achieved, ensuring the flatness of LST.

288 Tilt Angle Measurement

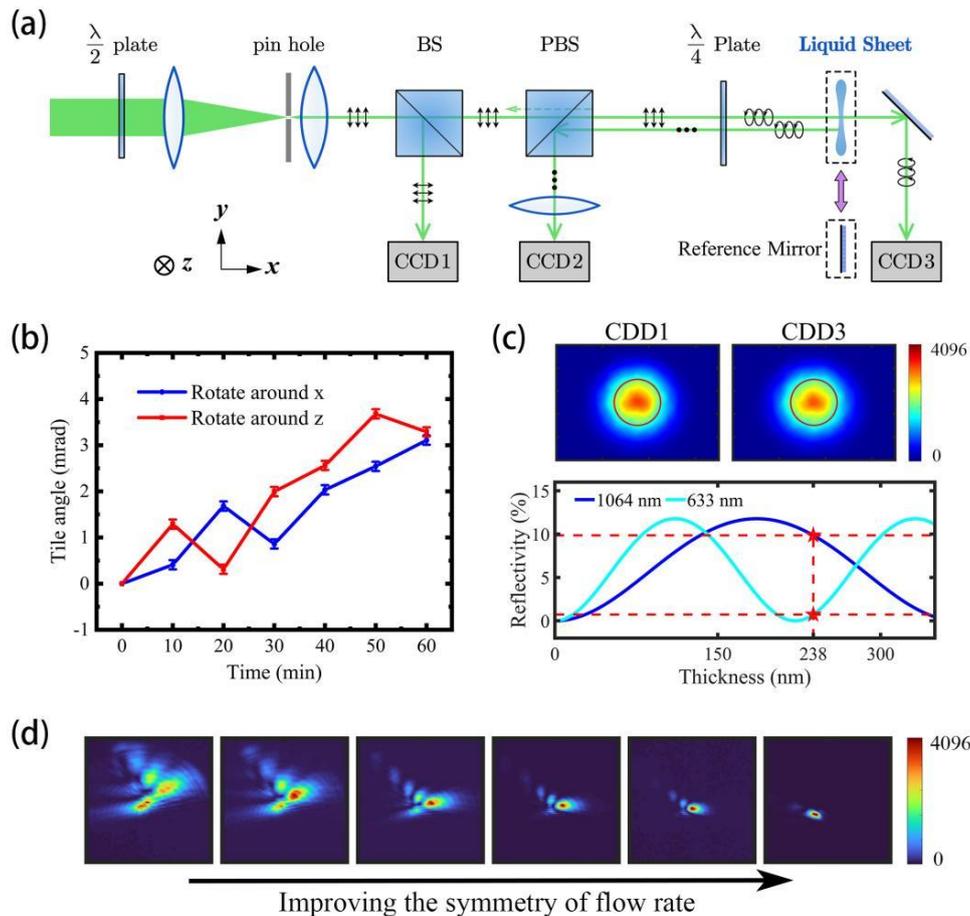
289 In laser ion acceleration, ions typically emit perpendicular to the surface of the
290 film. If the target normal is not precisely oriented towards the collimation entrance
291 of the ion energy spectrometer, the measured energy spectra may not be accurate.
292 Moreover, in applications where ions are collected and transported with magnets,
293 the correct orientation of the targets is crucial for designed beam transportation. The
294 tilt angles of solid film targets can be measured and adjusted mechanically, but this is
295 not feasible for self-supporting LSTs. Our diagnostic system includes a module to
296 measure the tilt angles of LSTs to ensure correct orientation.

297 As depicted in fig.5(a), a reference mirror first replaces the LST and reflects the
298 probe light to CCD2 via a polarization beam splitter (PBS) cube. The image spot on
299 CCD2 serves as the reference position. Thereafter, the reference mirror is switched to
300 the LST. The change in the position of the spot can be used to calculate the change in
301 the tilt angle based on the object-image relationship. In our system, the probe light is
302 initially rotated to s-polarized by a half-wave plate. After passing through PBS, light
303 reflected from the rear surface of PBS will not enter CCD2. The light reflected from
304 LST is converted to p-polarized after passing through a quarter-wave plate twice. It
305 will eventually enter CCD2. This compact design effectively eliminates the reflection
306 from the PBS, resulting in a good signal to noise ratio for the measurement. The
307 rightmost subgraph in fig.5(d) shows a typical reflection spot collected by CCD2. The
308 tilt angle of the LST can be calculated by the offset of the center of the reflection
309 spot with respect to the reference position.

310 The measurement accuracy can reach 0.2 mrad in our system, which is obtained
311 from the object-image relationship: the magnification of our system is 0.21, and the
312 distance between the object plane and LST is about 160 mm. The physical resolution
313 of the CCD is 1.34 μm . Assuming that the reflected light spot can be detected by
314 moving 5 pixels (in fact, using our algorithm to calculate the center of the light spot
315 can achieve an accuracy of 2-3 pixels), the resolution on the image plane is 6.7 μm ,
316 which is converted to 31.9 μm on the object plane. The field angle for the liquid
317 sheet is 0.2 mrad, which is sufficient for tilt angle diagnosis in laser ion acceleration.

318 Fig.5(b) shows an example measurement of the deviation of LST's tilt angle in 1
319 hour, the tilt angle of LST naturally jitters by several milliradians. If the tilt angle
320 deviates significantly due to some reason, this diagnosis allows us to detect and
321 recover it in time. The liquid sheet can be accurately regulated to point in the the

322 expected direction in two dimensions by adjusting the flow rate of the two pumps
 323 and the misalignment of collision.



324
 325 Fig.5 (a) Schematic diagram of tilt angle measurement, (b) The tilt angle change of LST within
 326 1 hour (the definition of the coordinate axis can be found in fig.1(b) and fig.5(a)), (c) The
 327 intensity integral of CCD1 and CCD3 for calculating absolute reflectivity of LST, (d) The
 328 change of spot collected by CCD2 with the improvement of flow symmetry.

329 CCD3 behind LST is used to image the sheet and the light spot on the sheet (the
 330 same CCD in fig.2(a)). This helps identify the exact location where the tilt angle of LST
 331 is measured, as the sheet may have different tilt angles at different positions due to
 332 distortion. CCD3 and CCD1 also form a spectroscopic path for measuring the
 333 thicknesses of LSTs thinner than 300 nm. Two co-propagating lasers with different
 334 wavelengths are irradiated on CCD1 and CCD3, as shown in fig.5(c), then the
 335 reflectivity of the film is calculated by measuring the integrated light intensity of
 336 CCD1 and CCD3 simultaneously to eliminate the impact of light source jitter. A pure
 337 and smooth film like a liquid sheet can exhibit minimal surface scattering, thus its
 338 absolute reflectance is measured. The theoretical reflectivity, calculated from
 339 equation(1), is illustrated by the two sine curves in fig.5(c), while the red pentagram

340 denotes the reflectivity measured at two distinct wavelengths. Analysis of this data
 341 suggests a film thickness of 238 nm, which is consistent with the results presented in
 342 the "Thickness Measurement" section at 240 nm. The resolution of this method for
 343 s-polarized light can be represented by the following equation:

$$344 \left(\frac{\partial R}{\partial h}\right)^{-1} = \frac{\lambda \left\{ 4 \cos^2 \theta \left[n^2(\lambda) - \sin^2 \theta \right] + \left[n^2(\lambda) - 1 \right]^2 \sin^2 \left[2\pi h \sqrt{n^2(\lambda) - \sin^2 \theta} / \lambda \right] \right\}^2}{8\pi \cos^2 \theta \left[n^2(\lambda) - \sin^2 \theta \right]^{\frac{3}{2}} \left[n^2(\lambda) - 1 \right]^2 \sin \left[4\pi h \sqrt{n^2(\lambda) - \sin^2 \theta} / \lambda \right]} \quad (3)$$

345 Assuming that $n = 1.43$, $\theta = 0$, $\lambda = 633\text{nm}$, when the measured thickness is 238
 346 nm, the result calculated from equation(3) is $\Delta h / \Delta R \approx 1175\text{nm}$. If $\Delta R = 0.1\%$ (this
 347 has been validated), then $\Delta h = 1.175\text{nm}$. This means that the thickness
 348 measurement result with error is 238 ± 1.2 nm.

349 Furthermore, CCD2 not only diagnoses the tilt angle of the sheet, but also
 350 provides more information about the flatness of the sheet near the measurement
 351 point. The distortion of the film due to the imbalance of the two colliding liquid flows
 352 would result in an aberrant image of the spot, similar to that caused by a non-flat
 353 mirror. By adjusting the jets, an ideal reflection spot can be achieved as illustrated in
 354 fig.5(d). As the symmetry of the flow rate improves, the center of the reflected spot
 355 gradually shifts, indicating that the tilt angle of LST changes. Compared to fig.4(e),
 356 which has a 3% asymmetry of flow rate, the method in fig.5(d) provides a preciser
 357 but more localized characterization of flatness with only 1% asymmetry of flow rate.

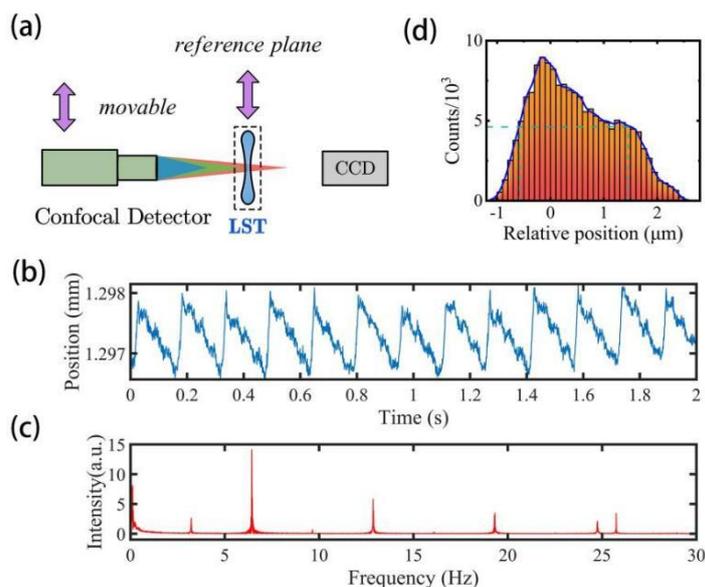
358 In summary, we have built an integrated module to measure the tilt angle of LST
 359 with an accuracy of ± 0.1 mrad. It also provides a quick and high-precision
 360 measurement of LST's thickness and local flatness.

361 Position Measurement

362 The results of laser ion acceleration closely relates to the relative positions of the
 363 laser focal spot and the target. Unlike solid films whose positions can be identified by
 364 defects or pollutants[26], a liquid sheet is transparent and flawless, thereby we
 365 utilized a confocal displacement detector (Micro-Epsilon, confocalDT 2421) to
 366 measure the position of LST.

367 The detector is arranged in the normal direction of LST and can be moved by a
 368 linear stage, as shown in fig.6(a). We first used a reference object to define this plane,
 369 and then used a CCD with very small depth of field (a few μm) to image it clearly.
 370 Main laser was focused by OAP to this plane as well. The confocal detector is then

371 used to measure the position of LST and make it coincide with the focusing plane.



372

373 Fig.6 (a)Schematic diagram of position measurement, (b)Time domain of LST's position
 374 (distance to the confocal detector), (c)Frequency domain of LST's vibration in (b),
 375 (d)Histogram of LST's relative position within 10 minutes.

376 Fig.6(b) shows the periodic vibration of LSTs position, which is detrimental to
 377 laser ion acceleration. Therefore, we analyzed it in the frequency domain, as shown
 378 in Fig.6(c), revealing that vibration can be attributed to specific frequencies. We
 379 found that 6Hz and its multiples are derived from pump plunger motion, while the
 380 rest include capillary vibration, platform inherent vibration, and so on. Fig.6(d) is a
 381 histogram of the position (or amplitude of vibration) of a LST within 10 minutes after
 382 optimization. Its FWHM is approximately 2 μm, which is smaller than the Rayleigh
 383 length of the focused laser. Some relevant discussions on how to suppress the LSTs'
 384 jitter can be found in our publication[9].

385 Conclusions and Perspectives

386 In this work, we present a comprehensive diagnostic system for real-time and
 387 in-situ characterization of liquid sheet targets in a vacuum chamber. Reflectance
 388 spectroscopy and interference fringes are employed to precisely measure LST
 389 thickness over the entire area, while their tilt angles and spatial positions are also
 390 measured simultaneously. We identified crucial parameters that impact the thickness
 391 of LSTs and investigated how flow asymmetry affects their flatness and tilt angles.
 392 This information enables precise and closed-loop control of LSTs, which is crucial for

393 laser-driven ion acceleration and other applications[27]-[32]. Furthermore, our
394 system and methods are also suitable for real-time, high-precision diagnosis of
395 transparent solid film targets.

396 **References**

397 [1] Higginson A, Gray R J, King M, et al. Near-100 MeV protons via a laser-driven
398 transparency-enhanced hybrid acceleration scheme[J]. *Nature communications*,
399 2018, 9(1): 724.

400 [2] Wang P, Gong Z, Lee S G, et al. Super-heavy ions acceleration driven by ultrashort
401 laser pulses at ultrahigh intensity[J]. *Physical Review X*, 2021, 11(2): 021049.

402 [3] Daido H, Nishiuchi M, Pirozhkov A S. Review of laser-driven ion sources and their
403 applications[J]. *Reports on progress in physics*, 2012, 75(5): 056401.

404 [4] Vozenin M-C, Bourhis J, Durante M. Towards clinical translation of flash
405 radiotherapy. *Nat Rev Clin Oncol* (2022) 19(12):791 – 803. doi:10.1038/s41571-022-
406 00697-z

407 [5] George K M, Morrison J T, Feister S, et al. High-repetition-rate (kHz) targets and
408 optics from liquid microjets for high-intensity laser – plasma interactions[J]. *High*
409 *Power Laser Science and Engineering*, 2019, 7.

410 [6] Morrison J T, Feister S, Frische K D, et al. MeV proton acceleration at kHz
411 repetition rate from ultra-intense laser liquid interaction[J]. *New Journal of Physics*,
412 2018, 20(2): 022001.

413 [7] Valdes P P, de Luis D, Hernandez J, et al. Implementation of a thin, flat water
414 target capable of high-repetition-rate MeV-range proton acceleration in a high-power
415 laser at the CLPU[J]. *Plasma Physics and Controlled Fusion*, 2022.

416 [8] Treffert F, Curry C B, Chou H G J, et al. High-repetition-rate, multi-MeV deuteron
417 acceleration from converging heavy water microjets at laser intensities of 1021
418 W/cm²[J]. *Applied Physics Letters*, 2022, 121(7): 074104.

419 [9] Cao Z, Peng Z, Shou Y, et al. Vibration and jitter of free-flowing thin liquid sheets
420 as target for high-repetition-rate laser-ion acceleration[J]. *Frontiers in Physics*, 2023,
421 11: 231.

422 [10] Ma WenJun, Liu Zhipeng, Wang Pengjie, et al. 2021 *Acta Physica Sinica* **70** 15.

423 [11] Li R, Ashgriz N. Characteristics of liquid sheets formed by two impinging jets[J].
424 *Physics of fluids*, 2006, 18(8): 087104.

425 [12] Born M, Wolf E. *Principles of optics: electromagnetic theory of propagation*,

- 426 interference and diffraction of light[M]. Elsevier, 2013.
- 427 [13] Watanabe A, Saito H, Ishida Y, et al. A new nozzle producing ultrathin liquid
428 sheets for femtosecond pulse dye lasers[J]. Optics Communications, 1989, 71(5):
429 301-304.
- 430 [14] Pano M R O, Delgado J M D. Effect of pre-impingement length and
431 misalignment in the hydrodynamics of multijet impingement atomization[J]. Physics
432 of Fluids, 2013, 25(1): 012105.
- 433 [15] Kashanj S, Kebriaee A. The effects of different jet velocities and axial
434 misalignment on the liquid sheet of two colliding jets[J]. Chemical Engineering
435 Science, 2019, 206: 235-248.
- 436 [16] Galinis G, Strucka J, Barnard J C T, et al. Micrometer-thickness liquid sheet jets
437 flowing in vacuum[J]. Review of Scientific Instruments, 2017, 88(8): 083117.
- 438 [17] Koralek J D, Kim J B, Brůža P, et al. Generation and characterization of ultrathin
439 free-flowing liquid sheets[J]. Nature communications, 2018, 9(1): 1353.
- 440 [18] Menzi S, Knopp G, Al Haddad A, et al. Generation and simple characterization of
441 flat, liquid jets[J]. Review of Scientific Instruments, 2020, 91(10): 105109.
- 442 [19] Hoffman D J, Van Driel T B, Kroll T, et al. Microfluidic liquid sheets as large-area
443 targets for high repetition XFELs[J]. Frontiers in molecular biosciences, 2022, 9:
444 1048932.
- 445 [20] Crissman C J, Mo M, Chen Z, et al. Sub-micron thick liquid sheets produced by
446 isotropically etched glass nozzles[J]. Lab on a Chip, 2022, 22(7): 1365-1373.
- 447 [21] Barnard J C T, Lee J P, Alexander O, et al. Delivery of stable ultra-thin liquid sheets
448 in vacuum for biochemical spectroscopy[J]. Frontiers in Molecular Biosciences, 2022,
449 9: 1044610.
- 450 [22] Goldstein R M, Zebker H A, Werner C L. Satellite radar interferometry:
451 Two-dimensional phase unwrapping[J]. Radio science, 1988, 23(4): 713-720.
- 452 [23] Abdul-Rahman H S, Gdeisat M A, Burton D R, et al. Fast and robust
453 three-dimensional best path phase unwrapping algorithm[J]. Applied optics, 2007,
454 46(26): 6623-6635.
- 455 [24] Chen M, Pukhov A, Yu T, et al. Enhanced collimated GeV monoenergetic ion
456 acceleration from a shaped foil target irradiated by a circularly polarized laser
457 pulse[J]. Physical review letters, 2009, 103(2): 024801.
- 458 [25] Hasson D, Peck R E. Thickness distribution in a sheet formed by impinging jets[J].
459 AIChE Journal, 1964, 10(5): 752-754.
- 460 [26] Shou Y, Wang D, Wang P, et al. Automated positioning of transparent targets

- 461 using defocusing method in a laser proton accelerator[J]. *Nuclear Instruments and*
462 *Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and*
463 *Associated Equipment*, 2019, 927: 236-239.
- 464 [27] Loh Z H, Doumy G, Arnold C, et al. Observation of the fastest chemical processes
465 in the radiolysis of water[J]. *Science*, 2020, 367(6474): 179-182.
- 466 [28] Yang J, Dettori R, Nunes J P F, et al. Direct observation of ultrafast hydrogen bond
467 strengthening in liquid water[J]. *Nature*, 2021, 596(7873): 531-535.
- 468 [29] Lin M F, Singh N, Liang S, et al. Imaging the short-lived hydroxyl-hydronium pair
469 in ionized liquid water[J]. *Science*, 2021, 374(6563): 92-95.
- 470 [30] Barnard J C T, Lee J P, Alexander O, et al. Delivery of stable ultra-thin liquid sheets
471 in vacuum for biochemical spectroscopy[J]. *Frontiers in Molecular Biosciences*, 2022:
472 1265.
- 473 [31] Yiwen E, Zhang L, Tsykin A, et al. Progress, challenges, and opportunities of
474 terahertz emission from liquids[J]. *JOSA B*, 2022, 39(3): A43-A51.
- 475 [32] Xia C L, Li Z L, Liu J Q, et al. Role of charge-resonance states in liquid high-order
476 harmonic generation[J]. *Physical Review A*, 2022, 105(1): 013115.