

## **Modification of laser-induced fluorescence spectrum by additional azimuthal Doppler effect in optical vortex beams**

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Laser-induced fluorescence method is a powerful tool to measure flow velocities of atomic and ionic species in a plasma. Optical vortex beams, which carry orbital angular momentum of light, have a potential to expand the capability of flow velocity measurements with laser-induced fluorescence method, because an atom moving in an optical vortex beam experiences additional Doppler effect in the azimuthal direction. The laser-induced fluorescence spectra obtained by substituting a commonly-used plane-wave-like beam with an optical vortex beam have been numerically evaluated. It is demonstrated that modification of the spectrum can be used to detect a fast flow perpendicular to the laser path, which is impossible to be observed by conventional method in principle. The use of standard deviation of the spectrum as an index of perpendicular flow velocity is also discussed.

## 1. Introduction

Flow velocities are important physical quantities to characterize weakly-ionized plasmas, and the laser-induced fluorescence (LIF) method<sup>1)</sup> is a powerful nonintrusive technique to measure the velocities of both ions and neutrals. A LIF spectrum obtained by scanning the frequency (i.e. wavelength) of narrow-band tunable laser across a resonant absorption line of the target atoms provides information on the velocity distribution function (VDF).<sup>2, 3)</sup> The broadening of VDF gives temperature, and the shift from the resonant absorption frequency for stationary particles gives mean flow velocity. It should be pointed out that the velocity obtained by the LIF method is only a component parallel to the laser path, because the technique is based on the longitudinal Doppler shift, which is proportional to the scalar product of the wave vector and the velocity vector.<sup>4)</sup> Thus, it can be said that the conventional LIF method is an essentially one-dimensional measurement. This one-dimensional nature is an advantage that interpretation of LIF spectrum can be performed straightforwardly; however, it is a disadvantage that no information on the velocity component perpendicular to the laser path is obtained in principle. In other words, the LIF method has no sensitivity in the direction perpendicular to the laser path. In many situations, the flow velocity toward an object, e.g. reactive ion species flowing onto a substrate in a plasma processing device, is desired to be measured. However, an ideal laser path to measure such velocity is perpendicular to the object and cannot be taken generally. If it is possible to make the LIF method sensitive to perpendicular movement of atoms, this limitation can be removed. The aim of this study is to explore the possibility of overcoming the limitation in LIF method by substituting a commonly-used lowest-order Hermite-Gaussian beam (HG<sub>00</sub>) with a Laguerre-Gaussian (LG) beam as known as *optical vortex*<sup>5-12)</sup>, which have not been used for LIF measurements in plasmas so far.

The LG modes are cylindrically-symmetric solutions of Helmholtz wave equation with a paraxial approximation in free space.<sup>13, 14)</sup> The LG<sub>*pl*</sub> beams are characterized by two integer mode numbers *p* and *l*. The radial intensity modulation is given by an associated Laguerre polynomial ( $L_p^l$ ). In this paper, *p* is set to zero for simplicity. The expression for LG beam intensity is given by the following equation:<sup>15)</sup>

$$I(r) = I_0 \left( \frac{2r^2}{w^2} \right)^{|l|} \left[ L_0^{|l|} \left( \frac{2r^2}{w^2} \right) \right]^2 \exp \left( -\frac{2r^2}{w^2} \right), \quad (1)$$

where *w* is a characteristic length which determines the spot size of the lowest-order mode. Figure 1 shows the beam intensity distributions of LG beams in which topological charges are *l* = 0, 1 and 10. When *l* = 0, the intensity pattern of LG<sub>00</sub> mode is a Gaussian function,

which is the same as HG<sub>00</sub> mode. In cases of  $l \neq 0$ , LG beams have a doughnuts shape in which the beam center has zero intensity. Moreover, the spot size increases with increasing the topological charge even when the parameter  $w$  has the same value.

A notable feature of LG beams is in their transverse phase structure, i.e. the phase varies in the azimuthal direction as  $\exp(il\phi)$ , where the angle  $\phi$  is the azimuthal coordinate in the cross section of the beam. Because of this  $\phi$ -dependence, any LG beam has a phase singularity on its central axis when  $l \neq 0$ . The equiphase surface is not a plane perpendicular to the beam axis but an  $l$ -intertwined helical structure, and the azimuthal mode number  $l$  is called as a topological charge. Due to this helical phase structure, the Poynting vector of an LG beam has the azimuthal component which produces an orbital angular momentum (OAM) even when the beam is linearly polarized.<sup>16-18)</sup> It should be emphasized that OAM of light is not associated with circular polarization in which a photon carries a spin angular momentum (SAM). This finding have led to various applications of OAM of light from optical manipulation<sup>19-21)</sup>, nanofabrication<sup>22, 23)</sup> to optical communication technology<sup>24, 25)</sup>.

An important property of an optical vortex beam for applying it to a laser measurement of plasmas is an additional Doppler effect in the azimuthal direction.<sup>26)</sup> An atom moving in an optical vortex beam experiences the azimuthal Doppler shift of resonant absorption frequency in addition to usual longitudinal Doppler shift. Therefore, a LIF spectrum obtained by using an optical vortex beam contains information on flow perpendicular to the optical laser path. In this paper, we numerically evaluate the shapes of LIF spectra obtained with optical vortex beams and discuss the feasibility to determine perpendicular flow velocity from the modification of a LIF spectrum. In the sections that follow, we first describe the resonant absorption condition for an atom modified by the additional azimuthal Doppler effect. A model for evaluating a LIF spectrum is then presented. Results clearly show the deformation of LIF spectrum especially in cases of fast flow crossing the laser path. Portions of this work were presented at the XXXIV International Conference on Phenomena in Ionized Gases in 2019 and published as an extended abstract.<sup>27)</sup> Discussions on the dependences of degree of modification, which is quantified by the standard deviation of obtained spectrum, on flow velocity and on spot size of the beam are included as new materials. Conclusions are given in the last section.

## 2. Evaluation method of LIF spectrum

### 2.1 Azimuthal Doppler shift in an optical vortex beam

The Doppler shift in a plane wave is longitudinal, i.e. it occurs only in the propagation

direction of light corresponding to wave vector  $\mathbf{k}$ . When the light is propagating in the  $z$ -direction, the Doppler shift of resonant absorption frequency of an atom moving at the velocity  $\mathbf{V}$  is given by  $\delta = -\mathbf{k} \cdot \mathbf{V} = -kV_z$ . However, the Doppler shift in an optical vortex beam is not one-dimensional due to the helical structure of the equiphase surface. The expression for the Doppler shift of resonant absorption frequency of an atom in an LG beam that propagates in  $z$ -direction with the topological charge  $l$  is given as follows <sup>26)</sup>:

$$\delta_{LG} = - \left[ k + \frac{kr^2}{2(z^2+z_R^2)} \left( \frac{2z^2}{(z^2+z_R^2)} - 1 \right) - \frac{(2p+|l|+1)z_R}{z^2+z_R^2} \right] V_z - \left( \frac{zkr}{z^2+z_R^2} \right) V_r - \left( \frac{l}{r} \right) V_\phi, \quad (2)$$

where  $V_z$ ,  $V_r$ ,  $V_\phi$  are the axial, radial and azimuthal velocity components of the atom,  $r$  is the distance from beam axis (phase singularity), and  $z_R$  is the Rayleigh length. Neglecting all the small terms coming from nonuniformity of the beam, we have two primary terms left in the expression as follows:

$$\delta_{LG} = -kV_z - \left( \frac{l}{r} \right) V_\phi. \quad (3)$$

The first term is the longitudinal Doppler shift utilized for traditional LIF method. Whereas, the second term is the additional azimuthal Doppler shift arising from the helical phase structure of optical vortex. The azimuthal Doppler shift is proportional to the topological charge  $l$  and inversely proportional to the distance  $r$  from the phase singularity. Therefore, in contrast to the plane wave case, the resonant absorption condition in an optical vortex beam depends on the position of the atom in a beam cross section except for the case when atoms are in rigid-body rotation about the beam axis. Figure 2 illustrates the position dependence of the azimuthal Doppler shift, where the parameter  $w = 5 \mu\text{m}$ , the perpendicular (beam-crossing) flow velocity to  $-x$ -direction (right to left) is  $U_x = 10 \text{ km/s}$ , the ion species is  $\text{Ar}^+$  of which temperature is  $0.1 \text{ eV}$ . The left panel shows the intensity distribution of  $\text{LG}_{01}$  beam. The right panels show LIF spectra obtained at the points A ( $\phi = 0$ ), B ( $\phi = \pi/2$ ), C ( $\phi = \pi$ ), and D ( $\phi = 3\pi/2$ ) on the circumference at  $r = 3.5 \mu\text{m}$ . The phase increases in the counter-clockwise direction. Since the azimuthal component of perpendicular flow velocity is  $U_\phi = -U_x \sin\phi$ , the blue shift of the spectrum is seen at B, and the red shift at D. No azimuthal Doppler shift is seen at A and C. This spatial dependence brings about a difference between LIF spectra obtained with plane-wave-like beams and LG beams.

## 2.2 Simplified model for constructing LIF spectrum

In this work, two simple assumptions have been made to calculate the LIF spectra as follows: (i) Observed (total) LIF spectrum is constructed by integrating a LIF contribution from each small segment on the beam cross section, (ii) LIF signal intensity is proportional

to the product of resonant absorption probability and the laser intensity at each point.

At first, we assume that the velocity distribution function (VDF) of ions is given by a shifted-Maxwellian,

$$F(V_r, V_\phi, V_z) = \left(\frac{1}{2\pi V_t^2}\right)^{\frac{3}{2}} \exp\left[-\frac{(V_r-U_r)^2+(V_\phi-U_\phi)^2+(V_z-U_z)^2}{2V_t^2}\right], \quad (4)$$

where  $V_t = \sqrt{k_B T_i/m_i}$  stands for the one-dimensional thermal velocity of ions,  $k_B$  the Boltzmann constant,  $m_i$  the ion mass, and  $U$  the flow velocity. By taking the Doppler shifted resonant absorption condition given by Eq. (3) into account, the absorption probability at a position  $(r, \phi)$  as a function of laser detuning  $\delta_{LG}$  is given as follows:

$$F(\delta_{LG}) = \left(\frac{1}{2\pi V_t^2}\right)^{\frac{1}{2}} \left(\frac{r^2}{l^2+k^2r^2}\right)^{\frac{1}{2}} \exp\left[-\frac{r^2}{2(l^2+k^2r^2)V_t^2} \left(\delta_{LG} + \frac{l}{r}U_\phi + kU_z\right)^2\right]. \quad (5)$$

In the case that  $U$  has only  $x$ -component for simplicity, the LIF contribution from a position  $(r, \phi)$  on the beam cross section is then given by

$$I_{\text{LIF}}(r, \phi) \propto \left(\frac{2r^2}{w^2}\right)^{|l|} \left[L_0^{|l|} \left(\frac{2r^2}{w^2}\right)\right]^2 \left(\frac{r^2}{l^2+k^2r^2}\right)^{\frac{1}{2}} \exp\left[-\frac{r^2}{2(l^2+k^2r^2)V_t^2} \left(\delta_{LG} - \frac{l}{r}U_x \sin\phi\right)^2 - \frac{2r^2}{w^2}\right]. \quad (6)$$

(See Appendix for detailed derivations.)

A LIF spectrum, which is to be observed in experiments, is obtained by numerically integrating the above expression on the  $r$ - $\phi$  plane. Note that the LIF spectrum loses information on the perpendicular flow direction due to integration, which is a disadvantage of this method.

### 3. Results and discussion

#### 3.1 Modification of LIF spectrum

Some examples of numerical calculation of LIF spectrum are shown in Fig. 3. Figure 3 (a) shows LIF spectra obtained with HG<sub>00</sub> plane wave (dotted line) and with LG<sub>01</sub> optical vortex (solid line) beams with the parameters  $w$  and  $U_x$  are 100  $\mu\text{m}$  and 1.5 km/s, respectively. Argon ion temperature is 0.1 eV. These values are relevant to our experimental devices HYPER-I<sup>28</sup>) and HYPER-II<sup>29</sup>), which are linear ECR plasma devices. There is no noticeable change in LIF spectrum due to azimuthal Doppler shift, because the shift is small compared with the Doppler broadening of 0.1 eV ions. Since the azimuthal Doppler shift is proportional to the topological charge and beam crossing velocity and is inversely proportional to the spot size of beam, we use LG<sub>010</sub> beam with  $w = 5 \mu\text{m}$  (spot size is about 30  $\mu\text{m}$ ) and assume much higher perpendicular velocity in Fig. 3 (b). Broadening of LIF spectrum is seen in the case of  $U_x = 5 \text{ km/s}$ , and remarkable modification is found in the case of  $U_x = 10 \text{ km/s}$ .

Quantification of the modification enables us to determine the perpendicular flow velocity from the LIF spectrum obtained with optical vortex beams. In the next subsection, we use the standard deviation as an index of the LIF spectrum modification.

### 3.2 Standard deviation of LIF spectrum

A higher-order moment is routinely used to characterize a velocity distribution function quantitatively. For example, the third-order moment (skewness) has been used to evaluate the asymmetry of LIF spectrum obtained in a plasma with inhomogeneity-induced radial flow in the HYPER-I device.<sup>30)</sup> In the present case, the LIF spectrum obtained with an optical vortex beam is symmetric. Therefore, as a first step, we utilize the standard deviation, which corresponds to the second-order moment, to evaluate the modification of LIF spectrum due to the azimuthal Doppler shift.

Figure 4 shows the standard deviation  $\sigma$  normalized by that of Doppler broadened spectrum for 0.1eV argon ions  $\sigma_0 = 0.735$  GHz, where the spot size parameter  $w = 5$   $\mu\text{m}$  and the topological charges are  $l = 1, 2, 3, 5$  and  $10$ . When the perpendicular flow velocity is slower than 5 km/s, which approximately corresponds to the ion sound speed of argon plasma with electron temperature of 10 eV, the value of  $\sigma/\sigma_0$  remains about unity. On the other hand, significant differences in  $\sigma/\sigma_0$  are seen for higher perpendicular flow velocities such as ion flow in the sheath of a negatively biased electrode. Therefore, the standard deviation of LIF spectrum can be used to determine beam crossing flow velocity especially in cases of high-speed flow. In experimental point of view,  $\sigma_0$  can be determined from the LIF spectrum obtained with a HG<sub>00</sub> beam on the same optical path. Since LG beams are produced by conversion of a HG<sub>00</sub> beam, another laser system is not necessary. Bypassing the conversion optics system enables us to use the original HG<sub>00</sub> beam for LIF measurement. The dependence of  $\sigma/\sigma_0$  on the spot size parameter  $w$  is shown in Fig. 5, where  $U_x = 10$  km/s. It is shown that the use of small spot size beam is required for having a significant difference in  $\sigma/\sigma_0$ . Even for  $l = 10$ , the maximum  $w$  for obtaining 10 % difference is 15  $\mu\text{m}$ , which corresponds to the spot size of about 90  $\mu\text{m}$ . It should be mentioned that this requirement is significantly altered by ion temperature, because the modification of LIF spectrum is produced by the competition between the azimuthal Doppler shift and the Doppler broadening due to ion temperature.

Through this paper, a drift-Maxwellian velocity distribution represented by Eq. (4) is assumed. Although a small asymmetry of velocity distribution gives only a slight

modification to the standard deviation, feasibility of separating contributions from the asymmetry and from the azimuthal Doppler shift is not a trivial problem. Validating application of the standard deviation method presented here to asymmetrical velocity distributions largely deviated from Maxwellian is our future work.

## 4. Conclusions

Substituting a commonly-used plane-wave-like beam (the lowest-order Hermite-Gaussian mode) with an optical vortex beam (Laguerre-Gaussian mode) has a potential to expand the capability of flow velocity measurement using conventional laser-induced fluorescence method by utilizing the azimuthal Doppler shift of resonant absorption frequency of atoms in an optical vortex beam. One of the advantages of this method is that the existing optical system can be used without alteration, because LG beams are solutions of Helmholtz wave equation in free space with paraxial approximation.

We have numerically evaluated the shapes of LIF spectra obtained by optical vortex beams with various topological charges and spot sizes. A significant modification of LIF spectrum has been obtained under the conditions that a high topological charge number of  $l = 10$ , a small spot size about  $30 \mu\text{m}$ , and a fast beam-crossing flow of  $10 \text{ km/s}$ . This modification can be used to detect a fast flow perpendicular to the laser path, which is not possible to detect by usual LIF method in principle. The use of standard deviation of LIF spectrum as an index of perpendicular flow velocity has been discussed, and its effectiveness has been demonstrated for fast flow cases. A proof-of-principle experiment for LIF method using optical vortex beams is left for our future work.

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## Appendix

For deriving Eq. (6), we first integrate the velocity distribution function given by Eq. (4) with respect to  $V_r$ . It gives

$$F(V_\phi, V_z) = \frac{1}{2\pi V_t^2} \exp \left[ -\frac{(V_\phi - U_\phi)^2 - (V_z - U_z)^2}{2V_t^2} \right]. \quad (\text{A}\cdot 1)$$

Using the resonant absorption condition Eq. (3),  $V_\phi$  in the above equation can be replaced by  $\delta_{LG}$  and  $V_z$ . By considering  $d\delta_{LG} = -\alpha dV_\phi$  ( $\alpha \equiv l/r$ ), we obtain

$$\begin{aligned} F(V_z, \delta_{LG}) &= \frac{1}{2\pi\alpha V_t^2} \exp \left[ -\frac{\left(\frac{kV_z + \delta_{LG} + U_\phi}{\alpha}\right)^2 - (V_z - U_z)^2}{2V_t^2} \right] \\ &= \frac{1}{2\pi\alpha V_t^2} \exp \left[ -\frac{1}{2V_t^2} \left\{ \frac{\alpha^2 + k^2}{\alpha^2} \left( V_z + \frac{k(\delta_{LG} + \alpha U_\phi) - \alpha^2 U_z}{\alpha^2 + k^2} \right)^2 + \frac{(\delta_{LG} + \alpha U_\phi + k U_z)^2}{\alpha^2 + k^2} \right\} \right]. \quad (\text{A}\cdot 2) \end{aligned}$$

The absorption probability function associated with Laguerre-Gaussian beams,  $F(\delta_{LG})$ , is then obtained by carrying out the integration with respect to  $V_z$ .

$$F(\delta_{LG}) = \frac{1}{2\pi(\alpha^2 + k^2)V_t^2} \exp \left[ -\frac{(\delta_{LG} + \alpha U_\phi + k U_z)^2}{2(\alpha^2 + k^2)V_t^2} \right]. \quad (\text{A}\cdot 3)$$

In the case of  $\alpha = 0$  ( $l = 0$ ), this probability function becomes the same form with that using a plane wave condition, and no additional Doppler effect perpendicular to  $\mathbf{k}$  appears. When the absorption of laser photons is in the linear regime, the LIF intensity at a laser detuning  $\delta_{LG}$  is proportional to both the absorption probability  $F(\delta_{LG})$  and the beam intensity  $I(r)$  given by Eq. (1). By using a Cartesian coordinates system with  $U_\phi = -U_x \sin \phi + U_y \cos \phi$ , we have

$$I_{\text{LIF}}(r, \phi) \propto \left(\frac{2r^2}{w^2}\right)^{|l|} \left[ L_0^{|l|} \left(\frac{2r^2}{w^2}\right) \right]^2 \left(\frac{1}{\alpha^2 + k^2}\right)^{\frac{1}{2}} \exp \left[ -\frac{(\delta_{LG} - \alpha U_x \sin \phi + \alpha U_y \cos \phi + k U_z)^2}{2(\alpha^2 + k^2)V_t^2} - \frac{2r^2}{w^2} \right]. \quad (\text{A}\cdot 4)$$

In a simple case of  $U_y = U_z = 0$ , Eq. (A·4) is equivalent to Eq. (6).

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## Figure Captions

**Fig. 1.** Beam intensity patterns of LG beams with the topological charges  $l = 0, 1$  and  $10$ . In cases of finite  $l$ , the beams have a doughnut shape. The spot size increases with increasing  $l$  even when the same value of  $w$  is used.

**Fig. 2.** The position dependence of the azimuthal Doppler shift, where  $w = 5 \mu\text{m}$  and  $U_x = -10 \text{ km/s}$ . LIF spectra obtained at the points A ( $\phi = 0$ ), B ( $\phi = \pi/2$ ), C ( $\phi = \pi$ ), and D ( $\phi = 3\pi/2$ ) on the circumference at  $r = 3.5 \mu\text{m}$  are shown in right panel.

**Fig. 3.** (a) Calculated LIF spectra for HG<sub>00</sub> (plane wave: dotted line) and LG<sub>01</sub> (optical vortex: solid line) beams. The spot size parameter  $w = 100 \mu\text{m}$ , and the perpendicular flow velocity  $U_x = 1.5 \text{ km/s}$ . There is no noticeable difference between two spectra. (b) Calculated LIF spectra for HG<sub>00</sub> (plane wave: dotted line) and two LG<sub>010</sub> (optical vortex: solid line and long and short dashed line) beams, where  $w = 5 \mu\text{m}$ ,  $U_x = 5 \text{ km/s}$  and  $10 \text{ km/s}$ . A modification of LIF spectrum due to the azimuthal Doppler shift is clearly seen.

**Fig. 4.** Dependence of normalized standard deviations of LIF spectra, which is obtained with  $w = 5 \mu\text{m}$  LG beams of which topological charges are  $l = 1, 2, 3, 5$  and  $10$ , on the perpendicular flow velocity. Increase of  $\sigma/\sigma_0$  can be seen in faster flow cases.

**Fig. 5.** Dependence of normalized standard deviations of LIF spectra on the spot size parameter  $w$ , where  $U_x = 10 \text{ km/s}$ . Small spot size is required to obtain significant difference.

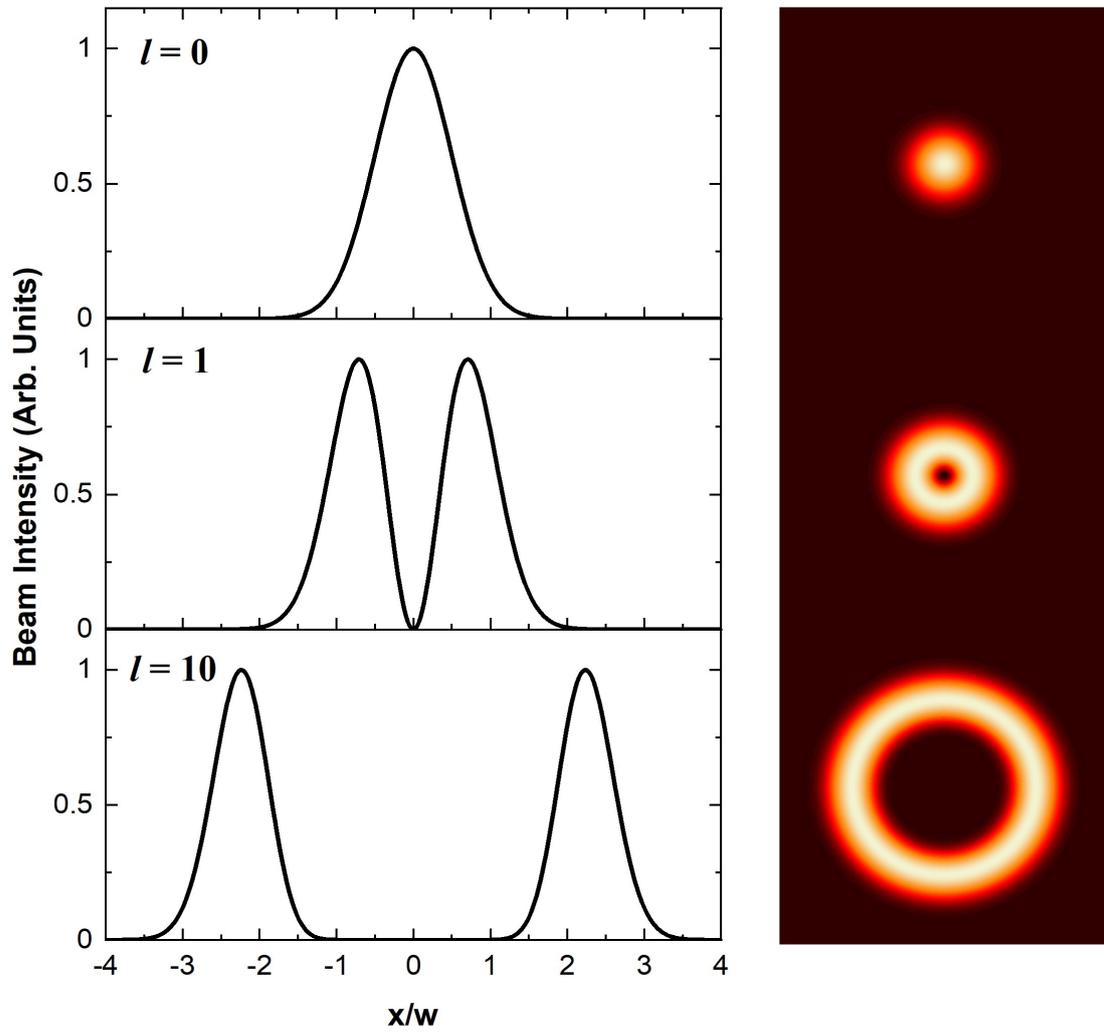


Fig. 1.

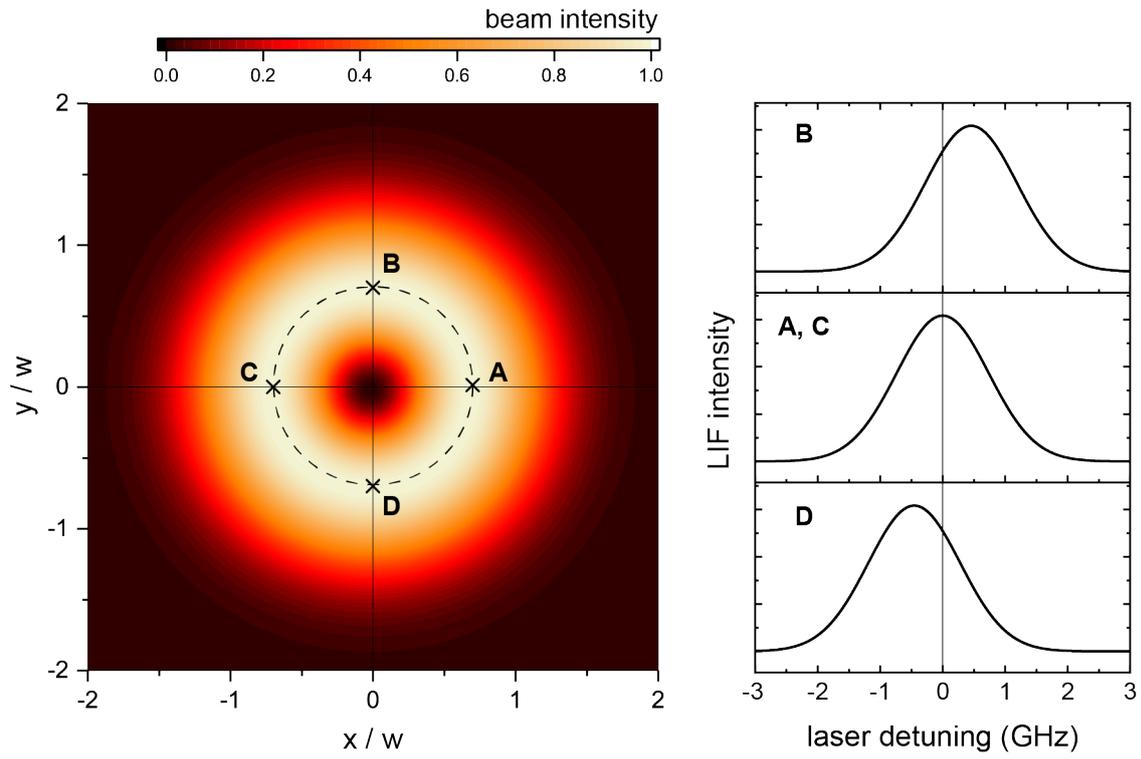


Fig. 2.

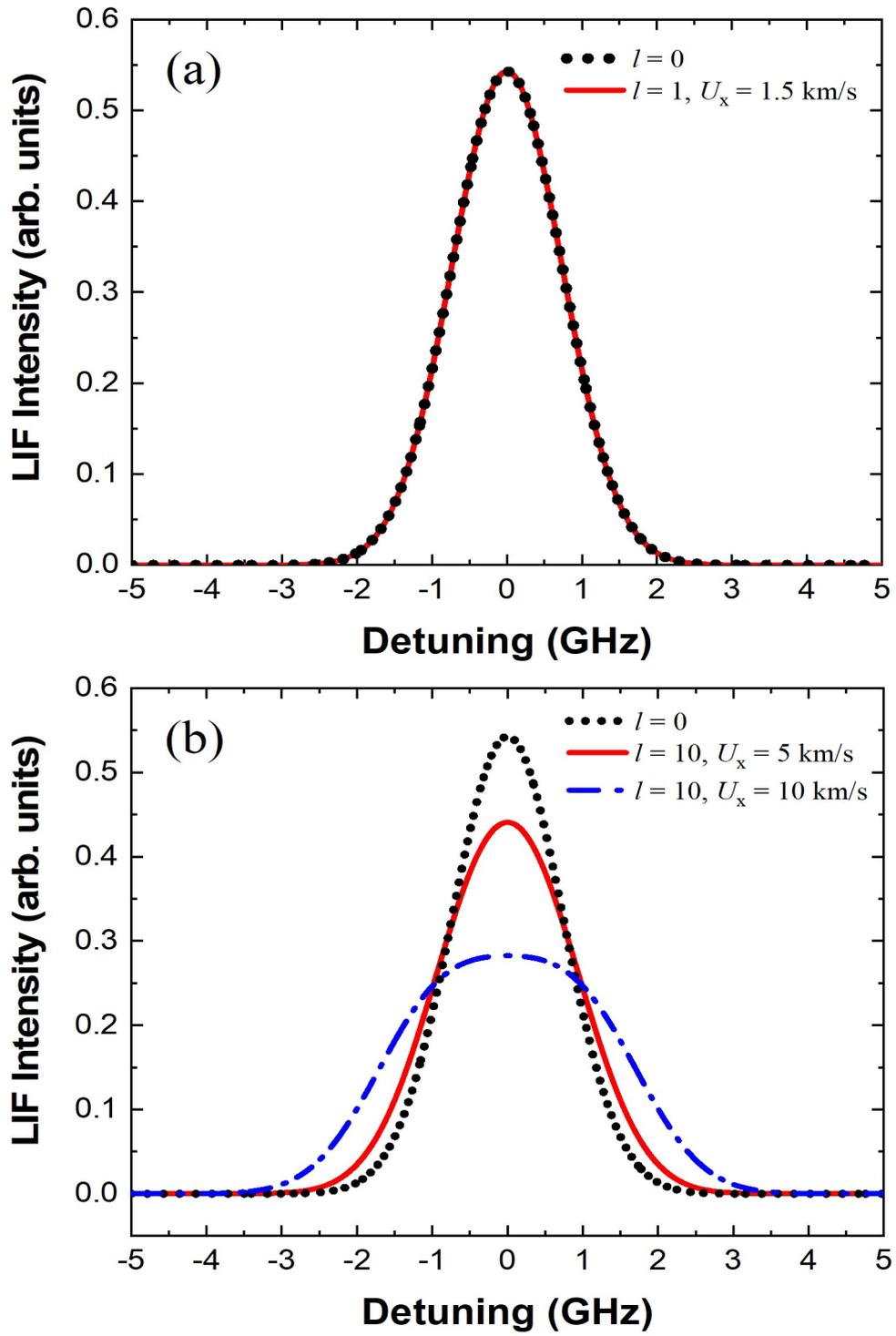


Fig. 3.

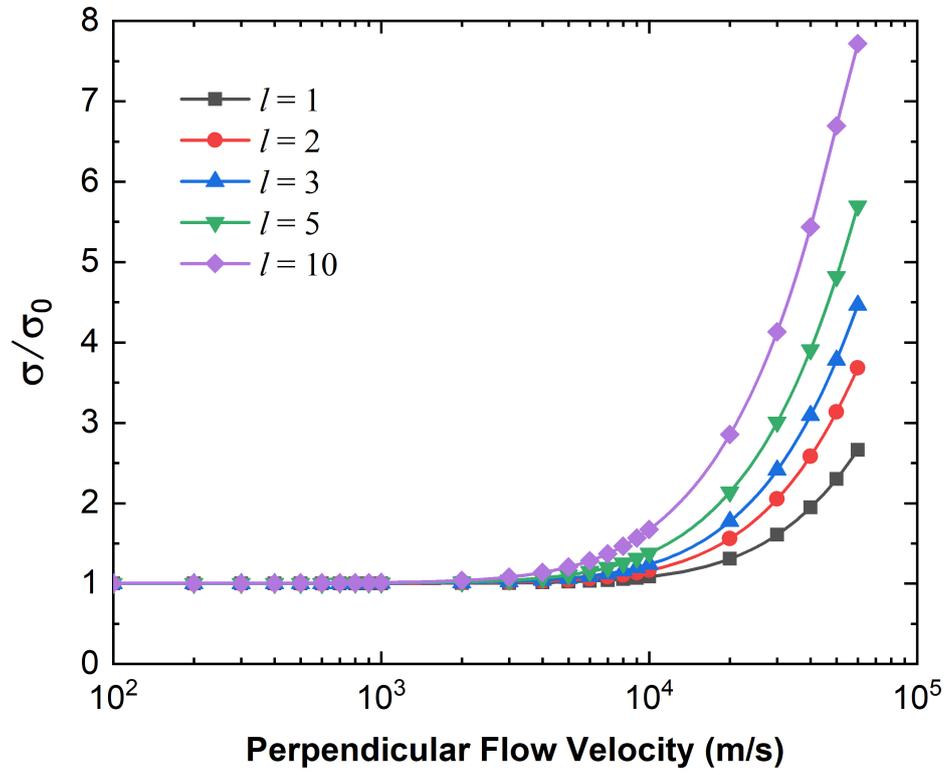


Fig. 4.

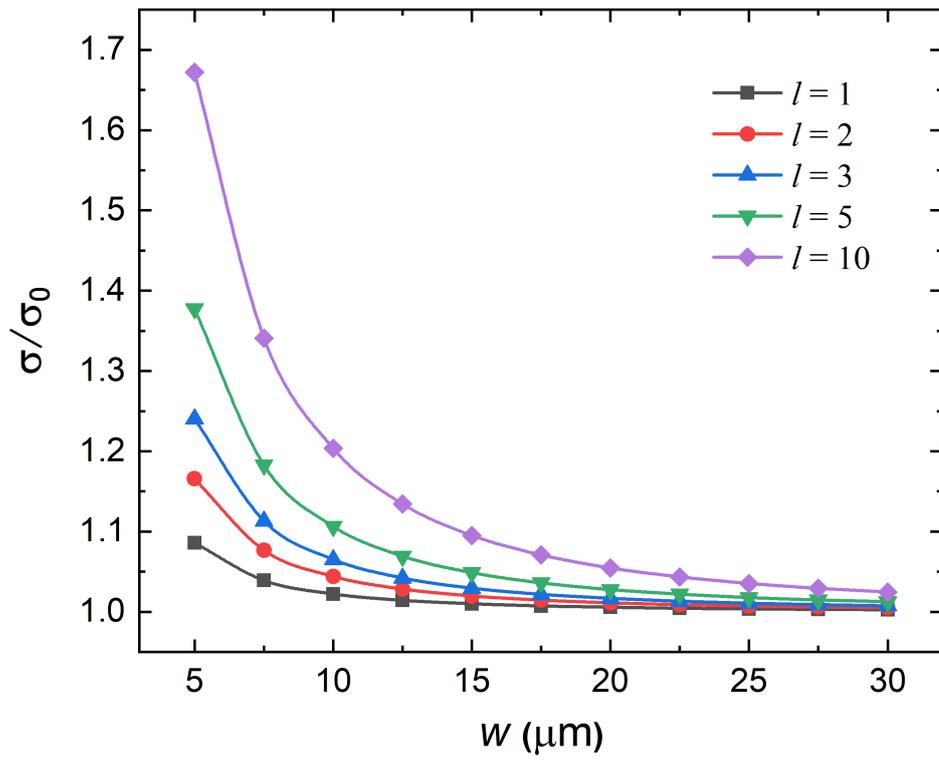


Fig. 5.