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Abstract. Atoms moving in an optical vortex beam are subjected to the azimuthal Doppler effect in addition to the usual longitudinal Doppler effect. This fact extends the capabilities of plasma flow measurement using laser-induced fluorescence to the direction perpendicular to the laser path by employing optical vortex beams. Furthermore, by assuming a uniform flow traversing the beam, the LIF spectrum undergoes deformation due to the spatial dependence of the resonant absorption condition. Preliminary experiments were performed for metastable argon ions in the vicinity of a negatively biased electrode immersed in a plasma. An increase in the standard deviation of the spectrum was observed when a negative voltage was applied to the electrode, which qualitatively agrees with our previous numerical study.

Keywords: optical vortex, Laguerre-Gaussian mode, laser-induced fluorescence method, flow velocity measurement, plasma diagnostics

1. Introduction

Laser spectroscopy, such as Thomson scattering, laser-induced fluorescence (LIF), and laser absorption spectroscopy (LAS), is a powerful tool for non-intrusive plasma diagnostics, widely used from magnetic confinement fusion plasma to industrial plasma application research. The TEM_{00} mode, which is a plane-wave-like Gaussian beam and an eigenmode of the laser cavity, has been used for those measurements. On the other hand, optical vortices, which are modes of light propagation with characteristic phase structures and carry orbital angular momentum, have attracted much attention in recent years [1,2,3]. Due to the presence of orbital angular momentum, optical vortices can exert a torque on microparticles, which has

led to applications such as the control of microparticles rotation by optical tweezers [4] and the formation of helical nanoneedle structures by laser ablation [5]. However, there are few examples of utilizing optical vortices for plasma measurement to the authors' knowledge. This paper proposes a possible application of optical vortices to plasma flow measurement.

LIF method with a narrow-band tunable laser is frequently used to measure the flows of ions and neutrals in plasmas. In the LIF method, an atom is excited by a laser beam with a wavelength corresponding to the energy difference between the two levels, and the light emitted upon de-excitation is measured. The LIF spectrum is obtained by sweeping the laser wavelength around the resonant absorption wavelength [6]. It should be noted that the LIF measurement using a plane-wave-like beam is, in principle, a one-dimensional measurement because it is based on the longitudinal Doppler effect. When the wavevector of the laser beam propagating in the z-direction is $\mathbf{k}$, and the velocity vector of an atom is $\mathbf{V}$, the Doppler shift of resonant absorption frequency, δ, is given by $\delta = -\mathbf{k} \cdot \mathbf{V} = -kV_z$. This equation shows that the flow perpendicular to the propagation direction has no contribution to the shift of the LIF spectrum. Although this one-dimensionality is advantageous in facilitating the analysis of the LIF spectrum, it is a disadvantage in limiting the information obtained from the spectrum.

We have been studying to extend the capability of the LIF method by changing the propagation mode of the laser light from TEM$_{00}$ to Laguerre-Gaussian (LG) modes [7], i.e., optical vortices. This paper describes the changes in the spectral shape in optical vortex LIF (OVLIF) measurement and reports the results of initial experiments for the proof of principle.

2. Optical vortex laser-induced fluorescence spectrum

The LG modes are cylindrically symmetric solutions of the Helmholtz equation in the paraxial approximation in free space. They are characterized by phase changes in the azimuthal direction on the cross-section perpendicular to the propagation direction. The intensity distribution of an optical vortex is donut-shaped because its optical axis is a phase singularity. When the phase change per revolution around the singularity point is $2\pi l$, l is called by the topological charge, an integer characterizing the phase structure.

Allen et al. showed in 1994 that an atom moving in an LG beam experiences an additional Doppler effect in the azimuthal direction [8]. Since the Doppler shift due to the non-uniformity of the beam is negligible in a collimated beam such as a laser, the principal terms for the Doppler shift, which is shown in ref. [8], can be simplified as follows.

$$\delta_{LG} = -kV_z - \left(\frac{l}{r}\right)V_\phi. \tag{1}$$

V_z, V_ϕ are the axial and azimuthal velocity components of the atom, k is the wavenumber, l is the topological charge, and r is the distance from the phase singularity at the center of the LG beam. The first term is the longitudinal Doppler shift used for the usual LIF method. The second term is the additional azimuthal Doppler shift that is specific to optical vortex beams.

The magnitude of the frequency shift is proportional to the topological charge and inversely proportional to the distance from the optical axis. Therefore, if there is a uniform flow traversing the LG beam, the magnitude of the second term will vary with position in the beam cross-section.

Consequently, the LIF spectrum is expected to be deformed because of the spatial dependence of the resonant absorption condition. As can be seen from (1), when $l = 1$ and the flow velocity is the same in the z and ϕ directions, the magnitude of the second term compared to the first term is very small, about the ratio of the wavelength to the beam size. Suppose it is possible to extract information about the flow-velocity vector of particles in the plasma from this slight azimuthal Doppler shift. In that case, we can develop a LIF method that is sensitive to the flow perpendicular to the beam propagation direction.

In our previous numerical study [9], we assumed for simplicity the following two conditions to evaluate the LIF spectrum observed in the experiment: (i) the LIF spectrum is the integral of the LIF signal contribution from a small segment on the beam cross-section, and (ii) the LIF signal intensity is proportional to the product of the resonance absorption probability and the laser intensity at each point. Assuming a Maxwell distribution as the distribution function and performing the integration in velocity space by taking the Doppler shift of the LG beam into account, the following expression was obtained.

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

Here, I_{LIF} is the LIF intensity at position (r, ϕ) on the beam cross-section, w is the characteristic length representing the spot size of the lowest-order mode, V_t is the thermal velocity, and U_x is the uniform flow in the x-direction. By numerically integrating (2), the LIF spectrum observed in experiments can be obtained. Although the integration loses the information on the flow direction, the magnitude of the flow velocity perpendicular to the beam path, which cannot be obtained by LIF using a plane-wave-like beam, is included. The numerical results showed that using an LG beam with a small spot size (typically on the order of 10 μm) and a large topological charge ($l = 10$ or more) for a fast flow of about 10 kms^{-1} results in significant deformation of the LIF spectrum. This deformation is due to the increase in spectral broadening caused by the azimuthal Doppler shift and can be evaluated using the standard deviation corresponding to the second-order moment as an index.

3. Experimental results and discussion

3.1. Experimental setup

A linear high-density plasma device HYPER-I [10] at the National Institute for Fusion Science (NIFS) was used for the experiment. A cylindrical vacuum chamber with a diameter of

30 cm and a length of 200 cm is surrounded by ten magnetic field coils to form a magnetic beach-type magnetic-field configuration. Electron Cyclotron Resonance (ECR) plasma is generated by injecting 2.45 GHz microwaves with right-handed circular polarization from the high-field side. In the present experiment, plasma was generated at an argon gas pressure of 3.0 mTorr and microwave power of 8 kW. The duration of plasma discharge was 45 s. The typical electron density and the electron temperature were 10^{12} cm^{-3} and 4 eV, respectively.

Figure 1 shows the experimental configuration. A 2.45 GHz microwave was injected through a tapered waveguide attached to the flange with a quartz window for vacuum-sealing. An electron cyclotron wave is excited in the plasma and fully absorbed in the ECR region in the middle of the vacuum chamber. A 50 mm diameter SUS disk electrode was placed at the center of the device, 117.5 cm from the microwave injection window. The ions can be accelerated by applying a negative voltage to this electrode. The LG beam was aligned so that it passed through in the very vicinity of the electrode, and the LIF is observed by the receiving optics installed above the device. The polarization of the LG beam was made to be linear in parallel with the magnetic field line to avoid Zeeman splitting. It should be pointed out that the polarization of LG beams can be selected freely because the orbital angular momentum of light is a different degree of freedom from the spin angular momentum that is related to the polarization. Since the fundamental transverse mode of the laser is TEM$_{00}$, it is necessary to convert the propagation mode to the LG mode by a certain method to realize LIF measurement with optical vortex beams in the laboratory. Spiral phase plates [11] with a thickness that varies in the azimuthal direction and q-plates [12], which are half-wave plates with the direction of the fast axis varying with the angle on the element, have often been used to generate optical vortices. However, the commercially available ones are only for low topological charges and cannot be applied to the optical vortex LIF method that requires a high topological charge. Therefore, we adopted a hologram method [13,14] using a spatial light modulator
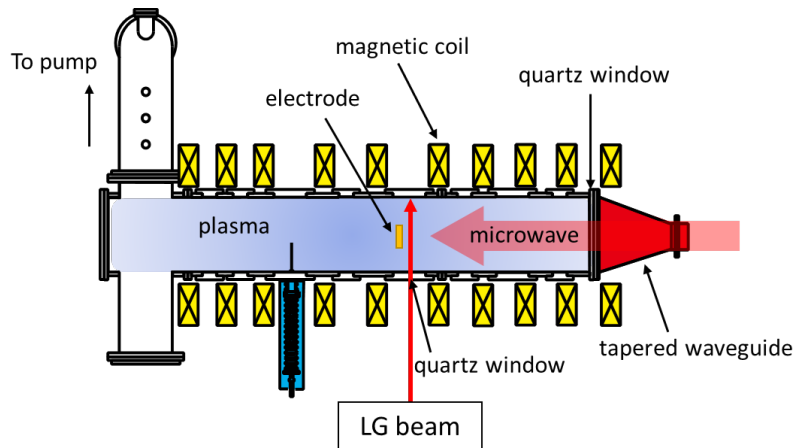

Figure 1: Experimental configuration. The LG beam is injected into the plasma through a quartz window on the side of the device.

(SLM) as the optical vortex generation method because of its high controllability of the topological charge.

Figure 2 shows the optical vortex generation and laser injection system. An external cavity tunable diode laser (TOPTICA, DL100) was used as the light source. The light at 668.6138 nm (448.3791 THz) that corresponds to the $3d^4\mathrm{F} \rightarrow 4p^4\mathrm{D}$ transition of argon metastable ions was amplified to 200 mW by a semiconductor optical amplifier (TOPTICA, BoosTA pro). After shaping the transverse mode using a single-mode fiber, the beam was modulated at 100 kHz by an electro-optic modulator (EOM) and converted into an $l = 10$ LG beam using a hologram method with SLM (Hamamatsu, LCOS-SLM X10468-07). The hologram used in this experiment, which was created by calculating the interference pattern of a plane wave and the $l = 10$ LG beam with a computer, is shown in Fig. 3. The LG beam is obtained as the first-order diffracted light by injecting the plane-wave-like beam into the SLM. Since the azimuthal Doppler shift is inversely proportional to the distance from the beam axis,
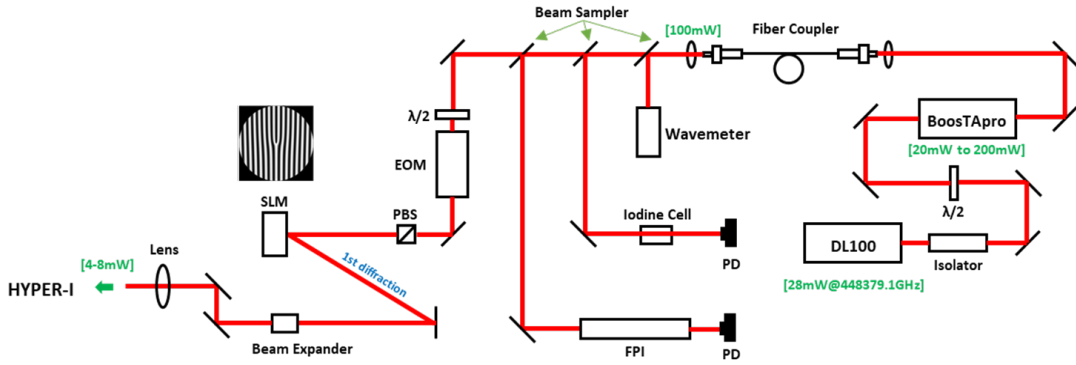

Figure 2: Layout of the optical system. The optical vortex beam was generated by converting the plane-wave-like beam using a hologram drawn on the SLM.
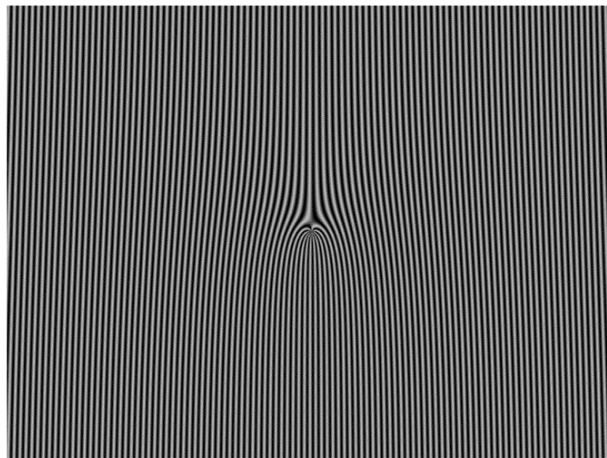

Figure 3: Computer generated hologram for $l = 10$ optical vortex beam.

the beam size should be reduced at the observation point. Therefore, a plano-convex lens was used to focus the beam in the very vicinity of the surface of the electrode.

The light-receiving system consisting of two plano-convex lenses and a multimode fiber was used to collect LIF, which is shown in Fig. 4. The 442.7 nm LIF corresponding to the $4p^4\text{D} \rightarrow 4s^4\text{P}$ transition was led to the photomultiplier tube (PMT) through an interference filter. The LIF signal detected using a lock-in amplifier (Stanford Research Systems, SR844) was recorded by a data logger (YOKOGAWA, ScopeCorder DL750P). The LIF spectrum was obtained by sweeping the laser frequency in a range of about ±6 GHz around the resonant absorption frequency of the stationary ion.
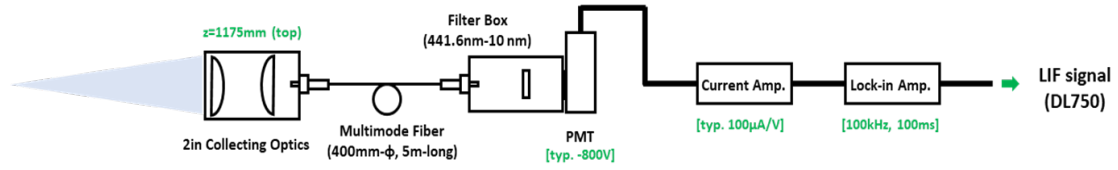

Figure 4: LIF receiving system.

3.2. Experimental results of OVLIF spectra

Figure 5 shows the OVLIF spectra obtained from the initial experiment. The vertical axis is the LIF intensity normalized to have an integral value of 1 in this range, and the horizontal axis is the detuning from the resonant absorption frequency for stationary metastable argon ions. Each spectrum is the average of five consecutive measurements under the same experimental conditions. The upper panel (a) shows the OVLIF spectrum measured with the electrode pulled down to avoid the effect of ion acceleration by the electrode's sheath. The shape of the OVLIF spectrum can be well approximated with a Gaussian distribution similar to that of the LIF spectrum measured with a plane-wave-like beam. In other words, no deformation due to the azimuthal Doppler shift can be seen. The middle panel (b) shows the OVLIF spectrum measured with the electrically-floating electrode, showing a slight broadening compared to the upper panel. The lower panel (c) shows the OVLIF spectrum when a negative voltage of -20 V is applied to the electrode, and there is an apparent deformation in the spectrum, although the signal to noise ratio (S/N) is reduced.

Taking moments is a method for quantitatively evaluating the deformation of a distribution. Table 1 shows each moment calculated from the spectra obtained in the experiment, where the defining equations for the first through fourth-order moments are as follows:

$$\mu_v = \int f(v)\mathrm{d}v, \tag{3}$$

$$\sigma^2 = \int (v - \mu_v)^2 f(v)\mathrm{d}v, \tag{4}$$

$$S = \frac{\int (v - \mu_v)^3 f(v) \mathrm{d}v}{\sigma^3}, \tag{5}$$

$$K = \frac{\int (v - \mu_v)^4 f(v) \mathrm{d}v}{\sigma^4}. \tag{6}$$
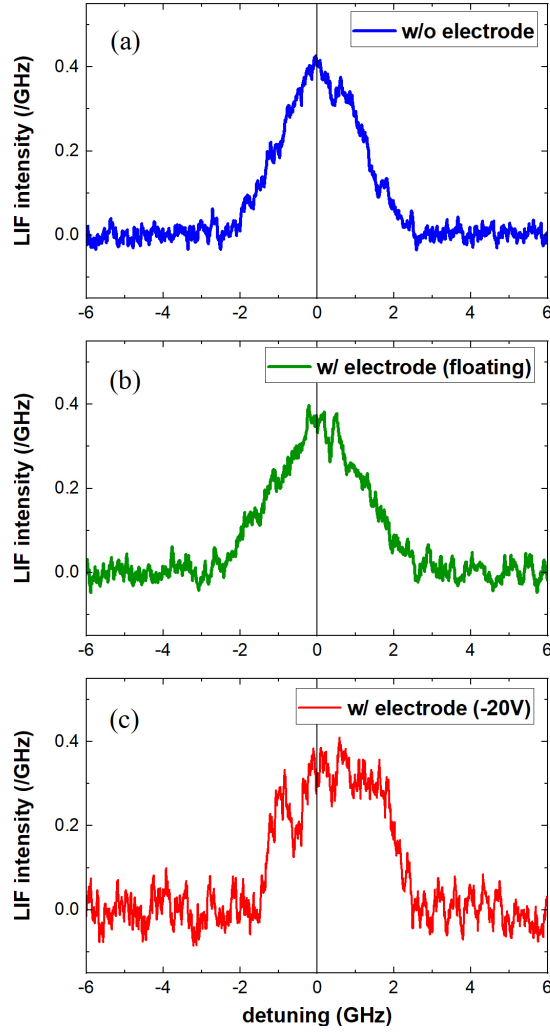

Figure 5: Examples of OVLIF spectra. (a) without electrode, (b) with electrically floating electrode, (c) with negatively (-20 V) biased electrode.

Table 1: Moments of experimentally obtained OVLIF spectra shown in Fig. 5

	(a)	(b)	(c)
1st, μ_v (flow $\parallel \boldsymbol{k}$)	0.11 GHz (73 m/s)	0.04 GHz (30 m/s)	0.33 GHz (226 m/s)

2nd, σ (Std. dev.)	0.88 GHz	1.11 GHz	1.28GHz
Skewness, S	0.049	0.003	0.017
Kurtosis, K	2.5	2.4	2.4

3.3. Discussion

The first-order moment (μ_v) shown in Table 1 corresponds to the flow parallel to the wavevector of the laser light. The values for (a) and (b) are reasonable because they are expected to be small due to the predominant flow along the magnetic field lines at the central part of ECR plasmas. As for the relatively large flow for (c), the cause has not clearly been understood, and it is hard to say definitively because of the poor S/N of the spectrum.

The third-order moments (S) for (a), (b), and (c) are all approximately zero, which indicates that the distributions are symmetric. The fourth-order moments (K) were almost the same values 2.4 for (a), (b), and (c). Since K takes a value of 3 for a Gaussian distribution, experimentally obtained OVLIF spectra have no heavy tail than the Gaussian distribution.

It should be noted that the standard deviations (σ) corresponding to the second-order moment increases when a negative potential is applied to the electrode. This result qualitatively agrees with our previous numerical study because the ions in the sheath/presheath region are accelerated by negative potential, resulting in a larger azimuthal Doppler shift. However, several factors still need to be considered to determine the flow velocity from the absolute value of the standard deviation. First, the spectra obtained experimentally have a finite offset, which leads to a non-negligible difference in the absolute value of the standard deviation from that of the ideal distribution. Second, our numerical calculations did not include the saturation effect of LIF. In the experiment, the beam was focused by a lens, and therefore the local energy density may have reached a saturation level. And third, there are other sources of broadening in the LIF spectrum other than the Doppler effect. For example, although the Zeeman effect can be neglected by aligning the polarization direction of the beam with the direction of the magnetic field lines, power broadening needs to be carefully evaluated.

To proceed to the detailed investigation described above, we need to improve the S/N of the OVLIF spectra under the presence of an azimuthal Doppler shift with a magnitude that is effective in deforming the spectrum shape. For example, since the azimuthal Doppler shift is inversely proportional to the distance from the singularity, the laser diameter must be reduced, which reduces the excitation volume and leads to less LIF intensity. Therefore, optimization of the beam diameter is necessary. Also, there may be a possibility of detecting the perpendicular flow using shifts rather than broadening by deliberately creating asymmetry in the beam intensity, such as asymmetric optical vortex beams [15]. These challenges remain for us

to address in the future.

4. Conclusion

Optical vortices have recently been attracted much attention in many research fields. However, there have been few examples of their application to plasma measurement. In this paper, we have reported the initial attempt to measure the ion flow velocity perpendicular to the laser path using the laser-induced fluorescence method using optical vortex beams. An increase in the standard deviation of the LIF spectra taken by an optical vortex beam has been observed by accelerating the ions in the direction perpendicular to the laser path using electrodes with a negative voltage applied, which qualitatively agrees with our previous numerical study. Further increase in S/N of the spectra and refinement of the numerical calculation lead to establishing the optical vortex LIF as a new plasma flow diagnostics.

Acknowledgment

References

[1] L. Allen, S. M. Barnett, M. J. Padgett: *Optical Angumar Momentum*, IOP Publishing, London, 2003.

[2] J. P. Torres, L. Torner: *Twisted Photons*, Wiley-VCH, Weinheim, Germany, 2011.

[3] D. L. Andrews, M. Babiker: *The Angular Momentum of Light*, Cambridge University Press, New York, 2013.

[4] D. G. Grier: A revolution in optical manipulation, *Nature*, 424 (2003), 810-816.

[5] T. Omatsu, K. Chujo, K. Miyamoto, M. Okida, K. Nakamura, N. Aoki, R. Morita, Metal microneedle fabrication using twisted light with spin, *Opt. Express*, 18:17 (2010), 17967-17973.

[6] W. Demtröder: *Laser Spectroscopy*, Splinger-Verlag, Berlin, 2003.

[7] A. E. Siegman: "Higher-order Gaussian modes" in *Lasers*, University Science Books, Palo Alto, 1986, 642-648.

[8] L. Allen, M. Babiker, W. L. Power: Azimuthal Doppler shift in light beams with orbital angular momentum, *Opt. Commun.*, 112:3-4 (1994), 141-144.

[9] S. Yoshimura, K. Terasaka, M. Aramaki: Modification of laser-induced fluorescence

spectrum by additional azimuthal Doppler effect in optical vortex beams, *Jpn. J. Appl. Phys.*, 59 (2020), SHHB04.

[10] S. Yoshimura, K. Terasaka, E. Tanaka, M. Aramaki, A. Okamoto, K. Nagaoka, M. Y. Tanaka: Exploration of spontaneous vortex formation and intermittent behavior in ECR plasmas: The HYPER-I experiments, *J. Plasma Phys.*, 81:2 (2015) 345810204.

[11] M. W. Beijersbergen, R. P. C. Coerwinkel, M. Kristensen, J. P. Woerdman: Helical-wavefront laser beams produced with a spiral phaseplate, *Opt. Commun.*, 112:5-6 (1994) 321-327.

[12] L. Marrucci, C. Manzo, D. Paparo: Optical Spin-to-Orbital Angular Momentum Conversion in Inhomogeneous Anisotropic Media, *Phys. Rev. Lett.*, 96:16 (2006) 163905.

[13] V. Yu. Bazhenov, M. V. Vasnetsov, M. S. Soskin: Laser beams with screw dislocations in their wavefront, *JETP Lett.*, 52:8 (1990) 429-431.

[14] A. V. Carpentier, H. Michinel, J. R. Salgueiro: Making optical vortices with computer-generated holograms, *Am. J. Phys.*, 76:10 (2008) 916-921.

[15] V. V. Kotlyar, A. A. Kovalev: Topological charge of asymmetric optical vortices, *Opt. Express*, 28:14 (2020) 20449-20460.