

DEVELOPMENT OF MEASUREMENT SYSTEM USING EVANESCENT WAVES FOR CHARACTERIZING COLLOIDAL LIQUIDS IN HEAT TRANSFER APPLICATIONS

K. SHIRAI, S. KAJI, T. KAWANAMI & S. HIRASAWA
Rokkodai 1-1, Nada, 657-8501, Kobe, Japan.

ABSTRACT

We report on the development of measurement system for characterizing physico-chemical properties of colloidal liquids used in heat transfer applications. In future thermal management, colloids consisting of micro- and nano-sized particles will play major roles in heat transfer for thermal storage and heat-transfer enhancement. In these applications, an important issue is the dispersion stability of colloidal particles. The functionality of the colloidal liquids becomes deteriorated when the particles aggregate and turn into sedimentation. The dispersion of colloidal liquid is maintained by the interaction of electrokinetic forces acting on the particles. The electrostatic state of the surface of a particle is represented by zeta potential, which represents the electrical potential difference between the particle surface and the surrounding. The zeta potential can be measured from the mobility of colloidal particles under electrophoresis. We use a pair of evanescent waves for measuring the zeta potential of colloidal particles. An evanescent wave propagates along an interface and exponentially attenuates away from it. The use of evanescent waves can achieve a spatial resolution smaller than a micrometer, which is not feasible with a conventional optical system whose resolution is bounded by diffraction limit. We describe the principle and design of the measurement system. A prototype measurement system was developed in the laboratory. We report on the development and performance of the system for characterizing colloidal particles for heat transfer applications.

Keywords: colloid, dispersion, evanescent wave, laser measurement, nanofluid, thermal storage, zeta potential

1 INTRODUCTION

Colloids are widely used in various applications. They consist of insoluble solute dispersed in solvent [1]. The quality of colloid is characterized by the dispersion stability, which describes the mixing degree of the solute and solvent without agglomeration or precipitation. There are various types of colloids with different phases of both solute and solvent. Among the different kinds of colloids, the one with both solute and solvent being in liquid phase is called emulsion. In thermal engineering field, an application of emulsion to the thermal storage is investigated using the latent heat of phase change material [2]. Another potential application of colloidal liquid is an enhancement of heat conduction with liquid containing nanometer-sized particles [3].

In colloid, solute particles are dispersed in the solvent with their electrical polarizations, which induced electrostatic interaction forces acting on the particles. The interaction forces include both attractive and repulsive forces. As long as the sum of the forces is dominated by the repulsive one, the suspension particles stably dispersed in the medium. Once the attractive force prevails against the repulsive one, the stability is hindered by aggregation and sedimentation. One of the keys for the dispersion stability is zeta potential, which is the electrical potential between the particle surfaces to the surrounding. If the particles have sufficiently large electrical charges, the resulting zeta potential becomes high and the particles remain stably dispersed. The zeta potential can be evaluated from the electrophoretic mobility of the solute particles at electrophoresis [1, 4]. The mobility can be evaluated by means of a micro-

scopic observation of particles motion and laser Doppler electrophoresis [5]. These optical methods are not applicable to highly dense colloids with little transparency. An ultrasonic method can measure colloids with high densities [6], but it has a difficulty in measuring emulsion with small density difference of solute and solvent.

The electrophoretic mobility of colloidal particle is measured with laser Doppler technique and hence the zeta potential is obtained. Since the penetration depth of an evanescent wave is in the range of 100 nm from a solid interface, the measurement volume can be placed in the vicinity of the interface, which enables measurements of highly dense colloid without the need of deep penetration of laser beams. The use of evanescent waves is commonly realized in total internal reflection fluorescence microscopes [7]. The use of evanescent wave can increase the spatial resolution of an optical measurement usually restricted to approximately half of the wavelength of light determined by the diffraction limit $\sim\lambda/NA$ (λ : wavelength, NA: numerical aperture). Especially, the use of an evanescent wave can increase the spatial resolution in the depth direction. Measurement of zeta potential based on the particle image velocimetry using evanescent waves was reported in the past [8].

Laser Doppler measurement using evanescent waves would further increase the measurement accuracy since it is based on frequency measurement. A laser Doppler velocimetry (LDV) using evanescent waves was once proposed in the past [9]. However, the system has never been used for the evaluation of zeta potential.

In the present study, we develop a new measurement system based on LDV using evanescent waves. The measurement volume is created with a pair of evanescent waves formed on a planar solid surface at total internal reflections. The very small penetration depth of evanescent waves is an advantage of increasing the spatial resolution and also of enabling the measurability of dense colloids. We describe the basic concept of the evanescent-wave-based LDV measurement system and introduce the design of the system. We built a prototype experimental system and investigated its performance.

2 THEORY

2.1 Zeta potential

Colloidal liquids consist of solvent with insoluble solute particles with their diameter ranging from 1 nm to 100 μm . In most cases, solute particles stay electrically charged and surrounded by ions charged with opposite sign as illustrated in Fig. 1. Ions with positive and negative signs are aligned along the interface between the particle and the surrounding, and they form an electric double layer. Individual particles move together with certain extent of the thickness of the double layer due to the viscosity. The outermost layer moving together with a particle is called slipping plane. The zeta potential is defined as the electrical potential difference between the slipping plane relative to the bulk fluid away from the layer. In colloidal liquid, both attractive and repulsive forces act between particles. The former one originates from the intermolecular force and the latter from osmotic pressure. When the attractive force is stronger than the repulsive force, the particles tend to agglomerate together. On the other hand, when the repulsive force is stronger than the attractive force, the particles remain dispersed. Since the repulsive force is proportional to the zeta potential, the dispersion stability can be known by measuring the zeta potential.

Zeta potential is evaluated by electrophoresis. When an electric field is induced to colloidal liquid, electrically charged colloidal particles move due to the electrostatic force. The electro-

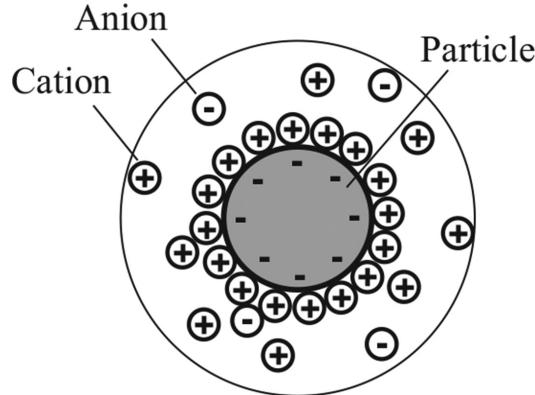


Figure 1: Schematic of an electrically charged solute particle in colloid.

phoretic mobility, i.e. flowing velocity of a particle v is proportional to the electric charge of the particle. Hence, the zeta potential can be derived from the electrophoretic mobility. The equation of Helmholtz–Smolkowski shows the relation between the electrical mobility and the zeta potential with an induced electric field E ,

$$\zeta = \frac{v\mu}{\varepsilon E}, \quad (1)$$

where ε and μ are dielectric constant and dynamic viscosity of the colloidal liquid. Hence, the zeta potential can be measured from the electrophoretic mobility.

2.2 Laser Doppler velocimetry

An LDV measures the velocity of a moving object based on the Doppler effect of light scattered by the object. A typical setup of an LDV used for applications in fluid mechanics is a differential configuration consisting of a pair of laser beams crossing at their beam waists as shown in Fig. 2. There exists a reference-beam configuration which also used in electrophoresis [1]. The beams create an optical interference at the crossing area, which serves as the measurement volume. The interference results in a fringe pattern parallel to the laser bisector plane. The fringe spacing d is described as

$$d = \frac{\lambda}{2 \sin \alpha}, \quad (2)$$

where λ and α being the wavelength and the half crossing angle, respectively. When a tracer particle passes through the measurement volume, it scatters the light and generates a modulated Doppler-bursting signal as shown in right side of Fig. 2. The beat frequency f of the signal is proportional to the velocity u perpendicular to the fringe bisector plane.

$$f = \frac{u}{d}. \quad (3)$$

Hence, the particle velocity is obtained by measuring the Doppler frequency. The spatial resolution is defined by the size of the measurement volume, which is determined by the

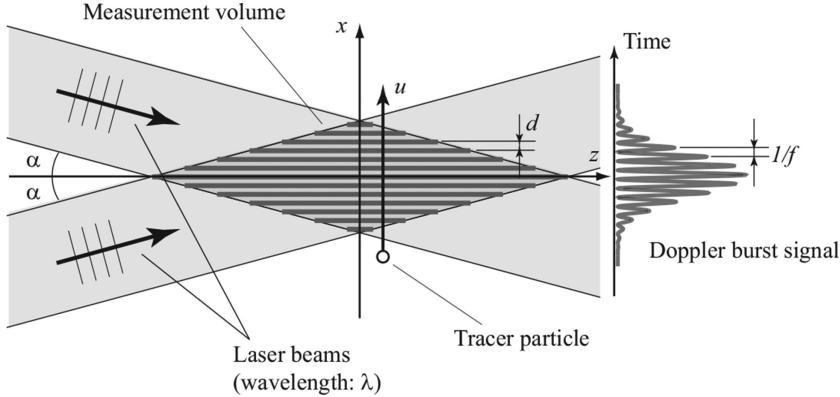


Figure 2: Principle of a differential laser Doppler velocimetry (LDV).

beam-waist diameter of the laser beams. The minimum focusable diameter of a laser beam is bounded by the diffraction originating from the wave nature of light. Hence, the spatial resolution cannot be increased beyond the diffraction limit, typically in the range of several tens of micrometers. The uncertainty of the frequency estimate is around $O(10^{-4})$. The resulting measurement uncertainty of an LDV is around $\sigma_u/u \approx 1\%$ due to the non-uniformity of the fringe spacings.

The zeta potential is evaluated based on the laser Doppler technique through eqn (1) by measuring the electrophoretic mobility of colloidal particles.

2.3 Evanescent wave

Now, we describe evanescent wave. Consider light propagation at a planar interface of two media with different refractive indices as shown in Fig. 3. Coordinates x and z are in the tangential and normal directions of the interface, respectively. The light propagates from the medium 1 with a refractive index n_1 toward medium 2 with an index n_2 . Assuming the incident medium has a higher value of the index compared to the other ($n_1 > n_2$), the refraction angle θ_2 is always larger than the incident one θ_1 . The light is partly refracted and partly reflected at the interface as shown in Fig. 3a. The refraction of the light is described by Snell's law.

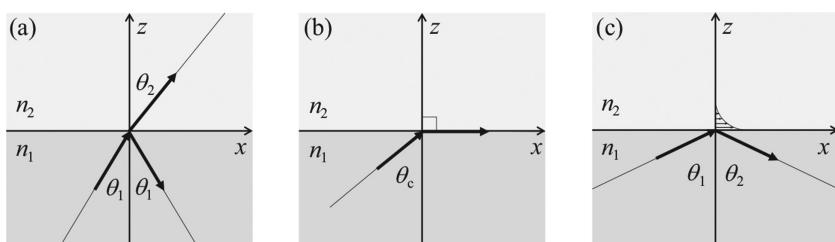


Figure 3: Refraction at a planar interface of two media with different refractive indices ($n_1 > n_2$), (a) ordinary refraction, (b) refraction at the critical angle, (c) total internal reflection.

$$n_1 \sin \theta_1 = n_2 \sin \theta_2. \quad (4)$$

With the increase of the incident angle, the refraction angle increases and reaches the right angle (Fig. 3b), at which the incident angle becomes

$$\theta_c = \sin^{-1}(n_2 / n_1). \quad (5)$$

This is the critical angle of incident, beyond which the incident light is totally reflected without refraction (see Fig. 3c). When an incident wave experiences a total internal reflection, an evanescent wave emerges at the interface on the other side of the incidence. Even though the incident wave is totally reflected back into the originating side with a higher refractive index, there is a penetration of wave into the other side at the interface. The electric field of the penetrating wave is the evanescent wave mathematically described as [10]

$$E_{ev} = A \exp(-j\beta z) \exp\{j(kx - \omega t)\}, \quad (6)$$

where A is the amplitude at the surface ($z=0$). The parameters j , t , ω denote the imaginary unit, time and the angular velocity of the wave, respectively. The constants in eqn (6) are

$$\beta = (\omega / c) \sqrt{(n_1 \sin \theta_1)^2 - n_2^2}, \quad (7)$$

$$k = (\omega / c) n_1 \sin \theta_1, \quad (8)$$

with c being the speed of light. These equations denote that the evanescent wave travels along the tangential direction of the interface between the two media. They also tell that the evanescent wave decays exponentially in the direction perpendicular to the interface. The inverse of β in eqn (7) is an indication of the penetration depth.

$$D = 1 / \beta = (\lambda_1 / 2\pi) / \sqrt{(n_1 \sin \theta_1)^2 - n_2^2}. \quad (9)$$

The penetration depth becomes approximately 160 nm in the case of $\lambda = 532$ nm (frequency doubled Nd: YAG), $n_1 = 1.52$ (for BK7), $n_2 = 1.33$ (for water) and $\theta = 70^\circ$ (cf. critical angle $\theta_c = 61^\circ$). Hence, the penetration depth is in the range of a few hundred nanometers at most.

The evanescent wave does not propagate into the medium 2, but once there is an object approaching to the interface in the side of the medium 2, the incident light starts to propagate into the medium following Snell's law.

3 EXPERIMENTAL SETUP

3.1 An LDV using evanescent waves

We described the concept of an evanescent-wave-based laser-Doppler velocimeter (EVLDV). The overview of the setup is shown in Fig. 4. It is a kind of Mach-Zender interferometer. A coherent laser beam is divided into two parts and finally guided into a glass plate, where the evanescent waves are generated for creating optical interference. The resulting Doppler beat frequency of the scattered lights from the two beams is obtained with heterodyne detection. The test section was designed to be filled with test liquid and two electrodes are placed inside (see Fig. 5). The two laser beams were incident from two prisms attached at the opposite sides on the glass plate. The incident beam angles were set beyond the critical angle so that they would create evanescent waves at the glass surface. The two evanescent waves form the measurement volume.

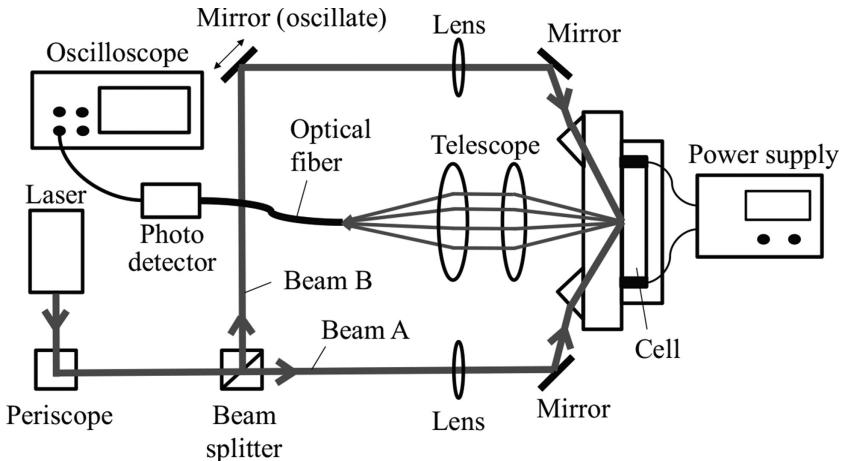


Figure 4: Schematic overview of the EVLDV measurement system.

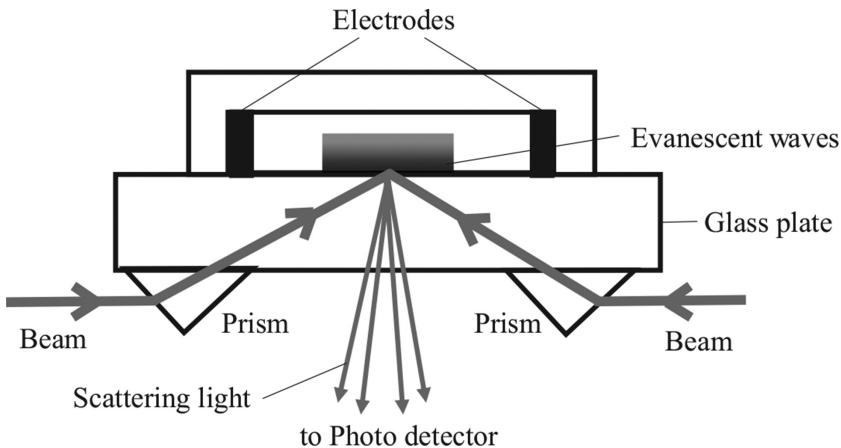


Figure 5: Schematic of the optical paths at the test section.

3.2 EVLDV system

The design of the optical system is shown in Figs 4 and 5 and the photograph of the setup is shown in Fig. 6. A laser source (wavelength: 532 nm, output power: 100 mW) emitted a single longitudinal mode beam, and it was guided through a periscope for beam steering. The beam was divided into two parts at a beam splitter cube (splitting ratio of 50:50). One of the beams indicated as Beam A in Fig. 4 propagated straight through an achromatic lens. The beam was reflected by a mirror and delivered into the test section through a right-angled prism. The other beam indicated as Beam B was reflected by another mirror and went through another achromatic lens. In some experiments, the mirror was oscillated with a piezo-electric actuator in the direction indicated in the figure. The beam was further reflected by another mirror and guided into the test section through another prism. The prisms and glass plates were all made of BK7. The thickness of the glass plate was 10 mm. The contact surfaces between the prisms and the glass plate were filled with highly viscous immersion oil. The incident beams were totally internally reflected, and evanescent waves were generated at

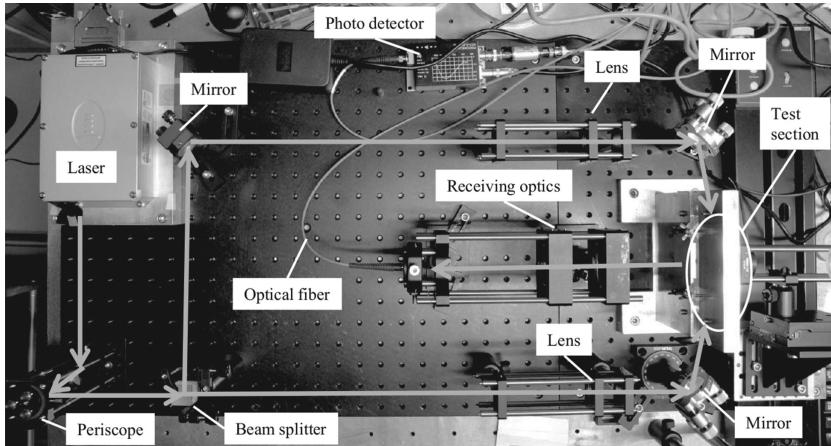


Figure 6: Photograph of the EVLDV system marked with laser beam paths.

the glass surface in the test section. The beams were adjusted by the reflection angles of the mirrors so that they cross at an identical location for forming a measurement volume. The incident angles of the beams were set asymmetric angles 71° and 75° at the total internal reflections in order to avoid possible damage to the laser source. The fringe spacings in the measurement volume were estimated to be 183 nm with a penetration depth of 129 nm. The test section was made water-tight and can be filled with test liquid. Two copper electrodes were attached in the section for electrophoresis. One side of the section was made of BK7 glass plate, on which evanescent waves were created. Other side of the section was made of poly-methyl methacrylate (PMMA). The scattered light was collected backward almost nearly sideward direction through a telescope consisting of a pair of achromatic lenses. The light was further guided into a photo detector through a multimode fiber. The light signals were converted into electrical ones and observed on an oscilloscope.

4 EXPERIMENT

4.1 Generation of evanescent waves

The first experiment was to generate evanescent waves at the glass surface. This experiment was carried out with test section detached. The incident laser beams into the prisms were adjusted so that their incident angles exceed the critical angles at the glass surface into the test section. We confirmed some scattering light on the surface under a condition that there was no refracted beam transmitted through the glass plate. The scattering light was considered to be caused by the evanescent waves.

4.2 Detection of evanescent waves

The second experiment was to detect the location of the evanescent waves generated at the glass surface. For the detection, a detection probe was developed with a combination of half-ball lens and rod lens as shown in Fig. 7a. They were glued together and a power sensor was attached to the backside of the rod lens. When the probe is not in contact with the glass surface, no light is detected at the sensor. Once the probe touches the glass surface, the eva-

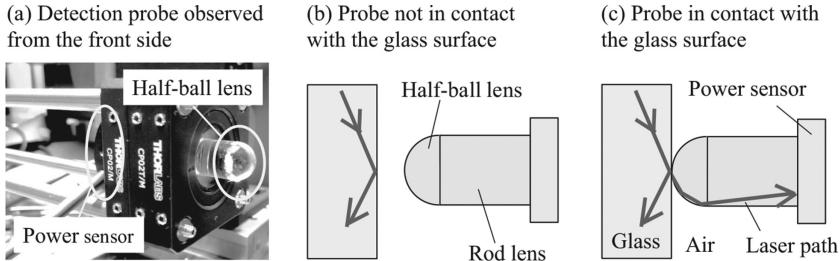


Figure 7: Detection method of evanescent waves using a self-developed probe.

nescent light starts to propagate into the half-ball lens. The light is transmitted into the rod lens after it is totally internally reflected several times on the curved surface of the lens. The transmitted light is finally detected by the power sensor placed at the end of the rod lens. No light is detected when the probe does not touch the location of the total internal reflection on the glass surface (Fig. 7b). Strong light is detected only when the probe precisely touches the location (Fig. 7c). By scanning the glass surface, the location of the total internal reflection and hence the evanescent light is identified with a precision of the beam radius at the glass surface. In an actual experiment, we measured maximum power of 1.0 mW during the scanning of the surface, while only 2.0 μ W was detected when the probe did not touch the surface. The 500 times difference of the optical power demonstrates the validity of the detection system. The method was used for the adjustment of the two beams so that the evanescent waves would create an interference at a desired location on the glass surface.

4.3 Measurement of Doppler signal

We tried to detect Doppler signals and to measure the Doppler frequencies. The two incident beams were adjusted using the detection method described in the previous subsection, the receiving optics were adjusted for detecting maximum power of the scattered lights. Scattered signals were detected when a scattering object was placed still against the glass surface

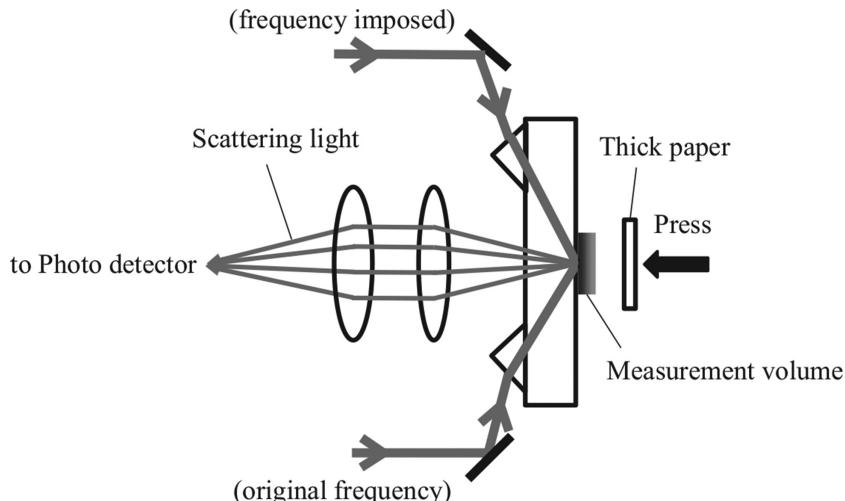


Figure 8: Schematic of the experiment of Doppler Signals and frequencies.

as shown in Fig. 8. The resulting signals in the range of several tens of mV were detected and their frequency spectra were observed. However, there was no frequency peak observed in the spectra except for the DC part. This was because the frequency contribution from the static scattering object is buried in the contribution from the DC part. No clear periodic oscillation or frequency peak was identified in the time signal and frequency domain even when the scattering object was moved tangentially along the glass surface. We oscillated one of the mirrors in the Beam B using a piezo electric actuator. In this case, a clear peak corresponding to the imposed oscillation frequency was observed in the spectra. However, there was no Doppler frequency observed for the scattering object in motion. We investigate possible causes for not being able to detect Doppler frequency corresponding to the scattering object.

5 CONCLUDING SUMMARY

We reported on the development a laser measurement system based on laser Doppler principle for the measurement of zeta potential of colloidal liquids for heat transfer applications. The concept and design of the EVLDV system was described with the theoretical consideration of evanescent waves. The optical system was built and evanescent waves were generated. A detection method of evanescent waves was developed and applied for the adjustment of the laser beams. An optical interference was created by overlapping the evanescent waves on the surface of a glass plate. Signals were detected using a scattering object placed in the measurement volume on the glass surface. Measurement feasibility of the EVLDV system was investigated.

ACKNOWLEDGEMENTS

The present work has been financially supported by the following organizations: Tanikawa fund promotion of thermal technology, Mikiya science and technology foundation, Kansai research foundation for technology promotion, TEPCO memorial foundation. We greatly acknowledge these supports.

REFERENCES

- [1] Cosgrove, T. (ed), *Colloid Science: Principles, Methods and Applications*, 2nd edn., John Wiley & Sons: Chichester, 2010.
- [2] Zhang, P., Ma, Z. & Wang, R., An overview of phase change material slurries: MPCs and CHS. *Renewable and Sustainable Energy Reviews*, **14**(2), pp. 598–614, 2010.
<http://dx.doi.org/10.1016/j.rser.2009.08.015>
- [3] Taylor, R., Coulombe, S., Otanicar, T., Phelan, P., Gunawan, A., Lv, W., Rosengarten, G., Prasher, R. & Tyagi, H., Small particles, big impacts: a review of the diverse applications of nanofluids. *Journal of Applied Physics*, **113**(011301), pp. 1–19, 2013.
<http://dx.doi.org/10.1063/1.4754271>
- [4] Probstein, R.F., *Physicochemical Hydrodynamics*, 2nd edn., John Wiley & Sons: Chichester, 2003.
- [5] Uzgiris, E.E., Laser doppler methods in electrophoresis. *Progress in Surface Science*, **10**, pp. 53–164, 1981.
[http://dx.doi.org/10.1016/0079-6816\(81\)90006-X](http://dx.doi.org/10.1016/0079-6816(81)90006-X)
- [6] O'Brien, R.W., Electro-acoustic effects in a dilute suspension of spherical particles. *Journal of Fluid Mechanics*, **190**, pp. 71–86, 1988.
<http://dx.doi.org/10.1017/S0022112088001211>
- [7] Axelrod, D., Total internal reflection fluorescence microscopy in cell biology. *Traffic*, **2**, pp. 764–774, 2001.
<http://dx.doi.org/10.1034/j.1600-0854.2001.21104.x>

- [8] Kazoe, Y. & Sato, Y., Measurements of electric double layer between electrolyte-glass interface by evanescent wave light illumination. *Proceeding of the International Symposia on Applicatona Laser Techniques to Fluid Mechanics*, eds R.J. Adrian, D.F.G. Durao, M.V. Heitor, K. Hishida & C. Tropea, Lisbon, pp. 1–11, 2004.
- [9] Yamada, J., Evanescent wave Doppler velocimetry for a wall's near field. *Applied Physics Letters*, **75**, pp. 1805–1806, 1999.
- [10] Hecht, E. (ed), *Optics*, 4th edn., Addison-Wesley: San Francisco, 2001.