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2007 Chinese Phys. Lett. 24 3358

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Observation of Diffusion Process of a Water Droplet Immersed in Protein Solution in Microgravity *

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(Received 3 September 2007)

Pure liquid-liquid diffusion driven by concentration gradients is hard to study in a normal gravity environment since convection and sedimentation also contribute to the mass transfer process. We employ a Mach-Zehnder interferometer to monitor the mass transfer process of a water droplet in EAFP protein solution under microgravity condition provided by the Satellite Shi Jian No 8. A series of the evolution charts of mass distribution during the diffusion process of the liquid droplet are presented and the relevant diffusion coefficient is determined.

PACS: 06.30.Dr

Mass transfer associated with diffusion takes place when the distribution of composition concentration in a solution is uneven. In normal gravity, the mass transfer due to diffusion is usually accompanied by mass transfer caused by convection and sedimentation. The diffusion coefficient of binary solutions is an important parameter in modelling studies of protein crystal growth. However, it is difficult to accurately measure diffusion coefficient with ground experiments because of the influence of convection and sedimentation caused by gravity. This situation impels scientists to conduct experiments on the diffusion coefficient of binary solutions in a microgravity environment. Among the related microgravity experiments, one is a space flight experiment, i.e., The Dynamics of Miscible Interfaces: A Space Flight Experiment, which is financed by NASA. Another is the Diffusion and Soret Coefficients Measurement for Improvement of Oil Recovery, which belongs to the Microgravity application promotion financed by ESA.

Scientists have developed research scope by measuring different liquid phase systems, and have made the experimental data more abundant. Theoretical coefficients of mass transfer, diffusion in two liquid phases, and diffusion of air and liquid mass transfer have been studied for a long time.^[1-4] The diffusion mechanism and the influence of concentration gradient to diffusion coefficient have also been discussed.^[5,6] Regarding methodology of measurement, in recent years, great research effort mainly focuses on measuring the diffusion process of liquid-liquid phase by interferometric method.^[7,8] However, the recording method of holographic interferometry and the difficulty in confirming the second exposure time make this technique limit its application to space experiments.

Rashidnia and Balasubramaniam^[9] designed a set

of common-path interferometer used in space experiment, and some experimental results in normal gravity have been obtained. The characteristic of this optical system is that the object light beam and the reference beam share a common path. The associated experimental facility is simple, non maladjusted, resisting vibration and therefore suitable for a space experiment. However, it has a lower measurement accuracy and sensitivity compared to two-path interferometer. In addition, its image processing is much more complicated in distinguishing information of object light beam and reference light beam because of the common path.

The German microgravity research centre has designed a two-wavelength Mach-Zehnder interferometer that is applicable in International Space Station (ISS). In ISS, the astronaut may operate the interferometer if the interferometer is maladjusted after the launching process. However, there is no astronaut available on the satellite in our present space experiment, an interferometer with ability to resist vibration and shock during the launching period of the rocket is desirable in order to study mass transfer in space. As we know, there has not been any actual application of Mach-Zehnder interferometers in space experiments reported in literature.

In this study, a specially designed Mach-Zehnder interferometer was used to monitor the mass transfer process of a water droplet in EAFP protein solution under microgravity condition provided by the Satellite Shi Jian No 8. A series of the evolution charts of mass distribution during the diffusion process of the liquid droplet are presented and the relevant diffusion coefficient is determined.

The Mach-Zehnder interferometer is a two-path optics measurement system, which has a high accu-

*Supported by the National Nature Science Foundation of China under Grant Nos 10432060 and 10672171, and the Knowledge Innovation Project of Chinese Academy of Sciences under Grant Nos KACX2-SW-02 and KSCX2-SW-322.

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racy. It has been used only in laboratory that is basically a vibration-free and shock-free environment. The facility used to observe the droplet diffusion on the Satellite Shi Jian No 8 is composed of four substantive parts, namely the Mach-Zehnder interferometer, image capturing system, the optical cells holding diffusion liquid samples, and the operation controlling system, as shown as in Fig.1. The Mach-Zehnder interferometer is sensitive and accurate, but it is also easily maladjusted by vibration, shock and temperature changing. To prevent the Mach-Zehnder interferometer from being maladjusted, some special anti-vibration measures were taken. For instance, a stainless steel board in the thickness of 14 mm has been used as optics soleplate, on which all three-dimensional adjuster are firmly locked after the optical modulating procedure. The mirrors are replaced by triangle prisms, and the beam splitters are replaced by splitter prisms in order to increase contacting area between optics glasses and optics soleplate. In addition, the optics glasses and the optics soleplate are firmly glued. The whole specially designed interferometer system, including optics system and image capturing system, has suffered from the serious tribulation during the rocket launching and in the space environment.

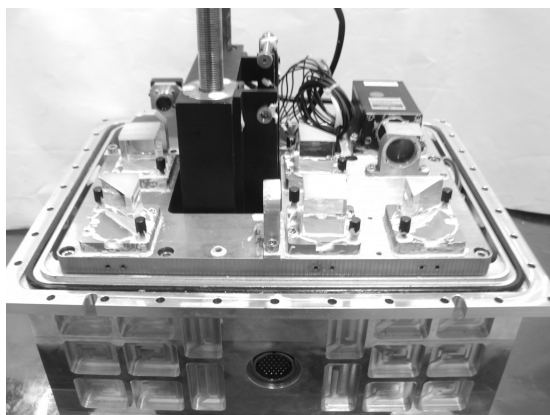


Fig. 1. Space experimental facility.

In order to calculate phase difference from interference fringes, Fourier transformation has been used to calculate the phase distributions of the metamorphic grating and the original grating. The change of refractive index $n(x, y)$ can be calculated by the relationship between phase difference and $n(x, y)$,

$$\frac{\Delta n(x, y) \cdot d}{\lambda} \cdot 2\pi = \Delta\phi(x, y), \quad (1)$$

$$\Delta n(x, y) = \frac{\Delta\phi(x, y) \cdot \lambda}{2\pi \cdot d}. \quad (2)$$

The local refractive index $n(x, y)$ is the original refractive n_0 plus the change of refractive index $n(x, y)$, i.e.

$$n(x, y) = n_0 + \Delta n(x, y). \quad (3)$$

The relationship between the refractive index and the concentration of EAFP protein solution was measured by WAY-15 ABBE refractometer in order to convert the refractive index field in the concentration field. The relative formula could be written as

$$C(\text{mg/mL}) = 1000.07n - 1325.5, \quad (4)$$

where n and $C(\text{mg/mL})$ are refractive index and concentration of EAFP protein solution respectively.

EAFP protein is a novel antifungal protein isolated from the bark of the tree *Ecommia Ulmoides* Oliver (Dozhong), which is a kind of Chinese herb medicine. It can be crystallized as a monoclinic form within several hours over wide pH range (4.5–7.8). Its molecular structure has been determined at atomic resolution (0.84 Å) by x-ray crystal diffraction technique. Measuring diffusion coefficient of EAFP solution will offer physics parameter for modelling studies of protein crystal growth.

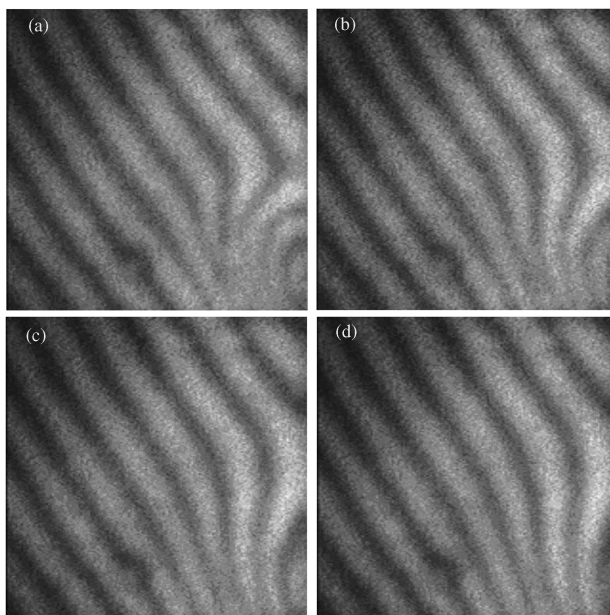


Fig. 2. Interference fringes in space experiment: (a) $t = 304.5$ s, (b) $t = 724.8$ s, (c) $t = 1087.3$ s, (d) $t = 1449.7$ s.

Diffusion process of a water droplet immersed in EAFP protein solution was observed in the space experiment on the Satellite Shi Jian No 8 launched in 2006, the space experiment images record the mass transfer process three hours at the recording rate of 5 images per second. Since the contribution from convection and sedimentation were excluded in microgravity, the mass transfer was caused only by a pure diffusion process. There is EAFP protein solution with the concentration of 10 mg/mL in a cell which is 6 mm × 6 mm × 1.5 mm in size. The thickness of the cell is 1.5 mm, which is the distance that the light travels through the cell. In order to calculate the concentration gradient of the diffusion process, a

water droplet with a much smaller size of 0.15 mm was injected into the cell at bottom.

Because of the much smaller water droplet and the rapid diffusion process, there is no obvious borderline

between the water droplet and the protein solution. However, the concentration gradient can be measured and calculated, and the diffusion mass transfer can be realized according to the changed interference fringes.

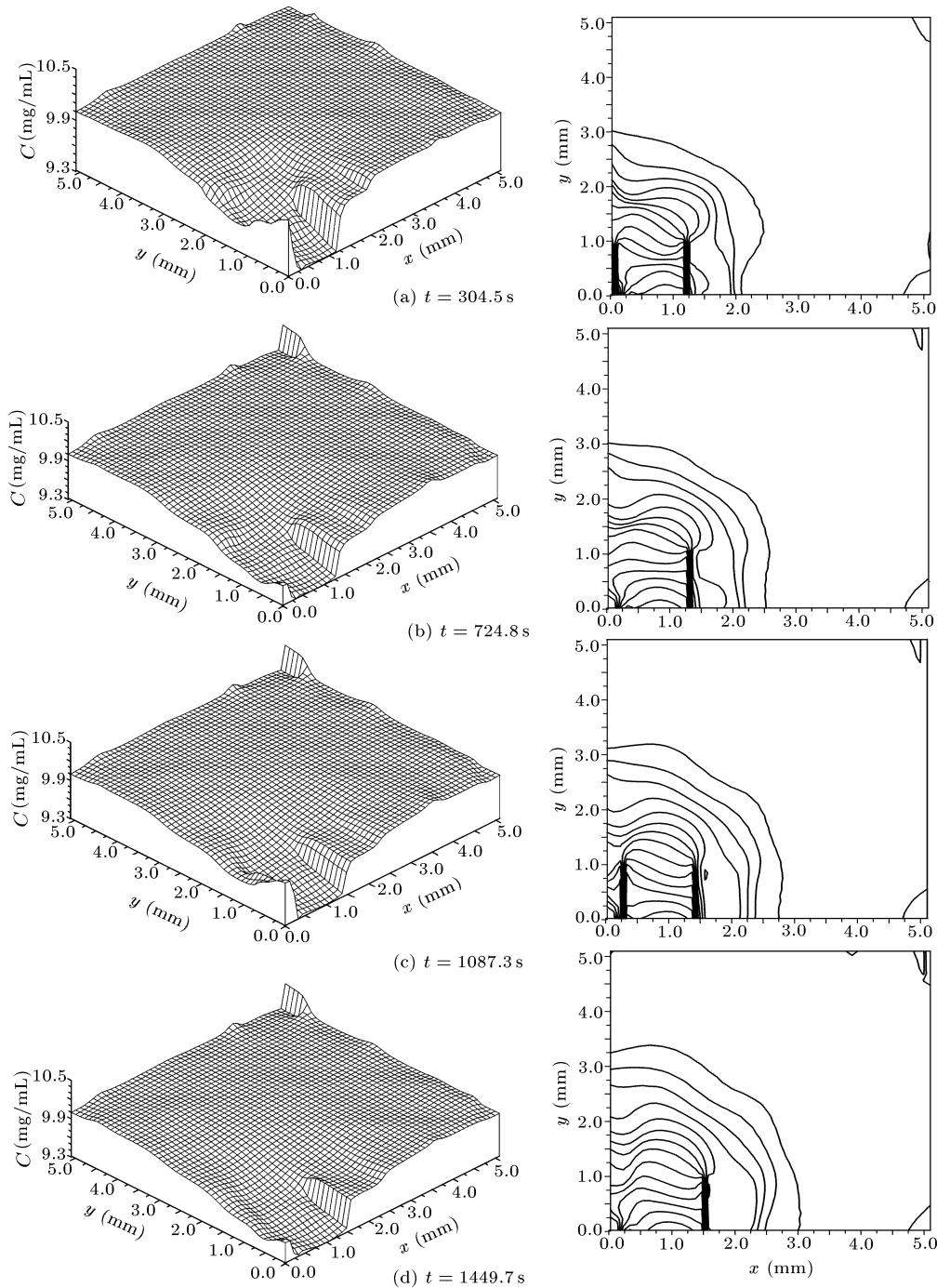


Fig. 3. Concentration distribution calculated from interference fringes.

In a microgravity environment, the diffusion process should be spherically symmetric, because the concentration gradients in all directions are equal. Figure 2 gives four photos of interference fringes taken at different times, which clearly show a steady diffusion

process, and the shape of droplet was well maintained for a very long period. The whole field concentration distribution at different times has been calculated from interference fringes according to the basic optics theory. The concentration distributions are shown

in Fig. 3. The concentration changes obviously, and the contour map shows concentric circles at the water droplet region. The concentration changes smoothly and gradually from the water drop. It is demonstrated that mass transfer in microgravity is a pure diffusion process.

Based on the analyses of the experiment data, concentration distributions along the x and y directions, at the time of $t = 1328.9\text{s}$ are presented in Fig. 4. Figures 3 and 4 show that the mass transfer in all the directions arrives at the distance of 4 mm from the water drop immersed and the diffusion speeds in all the directions are almost the same.

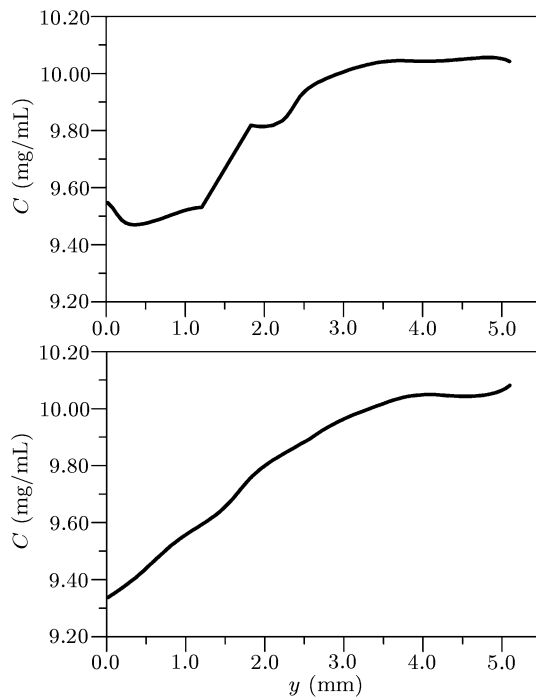


Fig. 4. Concentration distributions (a) along the x direction at $y = 0.7\text{ mm}$ and (b) along the y direction at $x = 0.7\text{ mm}$.

Because the space experiment opportunity is fairly scarce, only the results of one experiment have been obtained. However, the results we obtained can display the typical diffusion behaviour as the diffusion process under microgravity condition is theoretically induced by the concentration gradient.

Diffusion coefficient is calculated by the concentration distribution. Mass transfer in microgravity is a pure diffusion process, the diffusion speeds in all

the directions are almost the same. At the time from $t = 183.7\text{s}$ to $t = 606.5\text{s}$, the concentration distributions in the y direction at different x coordinates from 0.756 to 0.981 mm are approximately equal. In other words, for $x = 0.756\text{--}0.981\text{ mm}$ only the concentration gradient in the y direction exists, so the diffusion coefficient can be calculated by the one-dimensional method, i.e.

$$D = \frac{y^2}{4\Delta t} \{[\text{erfinv}(C_{t_2})]^{-2} - [\text{erfinv}(C_{t_1})]^{-2}\}, \quad (5)$$

where $C_t = \frac{[2C(x, t) - (C_1 + C_2)]}{(C_2 - C_1)}$, C_1 and C_2 are the concentration of EAFP solution and the water, respectively; $\Delta t = t_2 - t_1$, and erfinv is the inverse function of Gaussian error function. Therefore, the average diffusion coefficient is $2.0 \times 10^{-5} \text{ mm}^2/\text{s}$.

We demonstrate that a well-designed Mach-Zehnder interferometer is able to be used in space experiment facing the challenge from the serious tribulation of violent shock and vibration during the rocket launching period.

It is almost impossible to observe the diffusion process of a droplet on ground-based experiment because of the effects of sedimentation and convection caused by gravity. In contrary to this, the whole diffusion process of a water droplet in EAFP protein solution in microgravity was successfully observed by the Satellite Shi Jian No 8.

We present a series of the evolution charts of mass distribution during the long diffusion process to characterize the feature of diffusion of a liquid droplet in a solution and relevant diffusion coefficient determined by the experiment.

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