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Shape deformation of the gas-liquid interface of liquid oxygen in high-magnetic fields

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Shape deformation of the gas-liquid interface of liquid oxygen condensed in a glass tube in high magnetic fields is investigated by taking photos. It is found that there is a simple relationship between the deformation of the gas-liquid interface and the square of the applied magnetic field in a range of 0-~2 T, and that the shape of the interface changes tremendously from an ellipsoid into a sharp-pointed form with an increase in the field strength, breaking the linear relation. Regarding these experimental results, we carry out a discussion based on the phenomenological theory of a magnetic fluid.

Liquid oxygen is well known for its paramagnetism because the electronic ground state of this molecule is a triplet, and the study of this substance under a strong magnetic field is very interesting. Although some recent studies of the strong field effect on liquid oxygen[1,2] have been reported, no detailed study on the deformation of the gas-liquid interface with a magnetic field has yet to be published. As one of a series of studies on the physical properties of surfaces and phase nucleation, we made a direct observation of the shape of the gas-liquid interface of liquid oxygen in a glass tube suffering from a large deformation due to the magnetic field, and additionally investigated its detailed outlook by taking photos for the first time.

As a result of this study, it was found that the quasilinear relationship between the deformation of the gas-liquid interface and the square of the applied magnetic field in a range of 0-~2 T and that the shape of the interface changes from an ellipsoid into a sharp-pointed form with an increase in the field strength, breaking the linear relation. Regarding these experimental results, we carry out discussions based on the phenomenological theory of a magnetic fluid.[3]

The sample cell used in this experiment is a Pyrex glass tube with a 12-mm outside diameter and 10-mm inside diameter. In this cell, a sample gas with a purity of 99.995% was liquified by ~1 cm^3 after once passing through an LN_2 cold trap for further purification. In addition, this cell was placed nearly at the center of a split pair superconducting magnets provided with two holes with a 20-mm diameter in each of the X and Y directions. The magnet has a bore of 101.6 mm, maximum field strength of 7 T, and homogeneity of $\pm 0.1\%$ in the sphere of the 1-cm diameter at the center of the magnet.[4] The temperature of the sample cell was controlled to a precision better than ± 0.1 K by regulating the vapor pressure of the liquid nitrogen in a glass Dewar mounted on the outside of the cell with a manostat. In this experiment, light from a mercury lamp was introduced from outside of the cryostat to the sample chamber through the holes in the magnet to magnify and observe the appearance of the interface with a telescopic lens, and photos were taken. In this case, a 546-nm filter was used for the light of the Hg lamp to obtain clear images.[5]

Figure 1 shows photos of the deformed gas-liquid interface of oxygen at a temperature of 65.3 K in a magnetic field up to 5 T. These photos were taken in a homogeneous magnetic field applied upward. Next, to explain each portion of these photos appropriately, illustrations are used. Figure 2 is an illustration of the gas-liquid interface

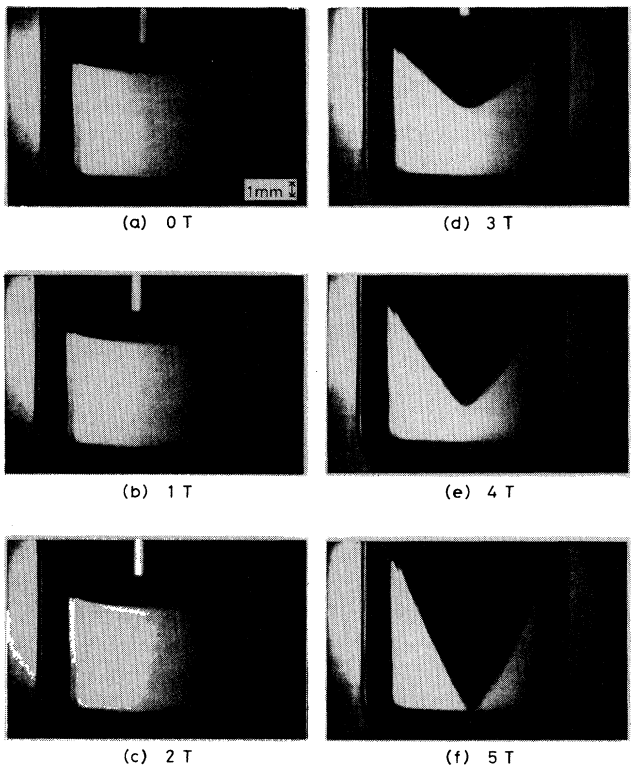

FIG. 1. Photos of the deformed gas-liquid interface of liquid oxygen at the temperature of 65.3 K in the magnetic field up to 5 T. In each photo, a large bright part in the central portion and a dark part at the upper portion correspond to the sample liquid and the saturated vapor of oxygen, respectively. In the gaseous portion, there is a bright vertical bar in (a)-(d). The bar appears for some reasons of optical apparatus used, and is not as important for the observations of the gas-liquid interface.

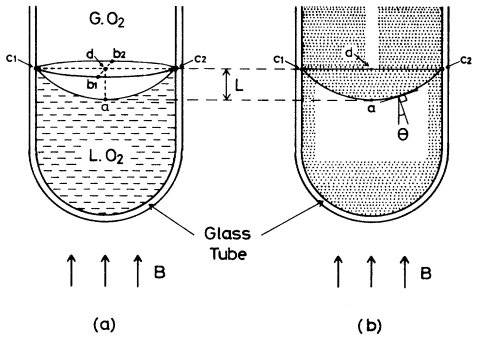

FIG. 2. An illustration of the gas-liquid interface in the applied magnetic field of 2 T, corresponding to the photo in Fig. 1(c).

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The actual gas-liquid interface has a three-dimensional (3D) shape as shown in Fig. 2(a), but the interface appears to be 2D as shown in Fig. 2(b) when the photo is taken just from the side. The line c_1-a-c_2 in Fig. 2(b) shows the gas-liquid interface, and point a is its lowest point. Also, points b_1, b_2, c_1, and c_2 in Fig. 2(a) lie on a horizontal plane, and show the highest portion where the sample liquid has risen along the inner wall of the cell due to surface tension. The photo in Fig. 1(c) has a bright part in the central portion which is the sample liquid and a dark part at the upper portion which is the gas. This darkness is caused by the difference in the refraction index between the gas and liquid. Also, in the gaseous portion, the part above point d, located directly above point a in Fig. 2, appears partially bright. In addition, both sides and the vicinity of the bottom of the cell show up dark in the photo because the sample cell is made with a glass cylinder. However, these areas of partial brightness and darkness in the photos have no effect on this experiment.

From a series of photos in Fig. 1, it is found that the gas-liquid interface clearly extends in the direction of the magnetic field as the field strength is increased from zero in the direction of the sample cell axis. Particularly above 3 T, the extension is found to be rather distinct, and also the shape of the gas-liquid interface changes from an ellipsoidal to a sharp-pointed one. When the magnetic field was raised and applied above $\sim$6 T (omitted in Fig. 1), it was observed that the sample liquid became detached from the cell bottom and completely adhered to the inner wall. These phenomena were similarly observed regardless of the applied magnetic field in Fig. 2, upwards or downwards. Also, the response time of the interface deformation with respect to the magnetic field was very short, no hysteresis of the deformation was found against a variation of the magnetic field, and reproducibility was very good, too.

Next, to quantitatively analyze the deformation of the gas-liquid interface of oxygen with the magnetic field, the length of a characteristic portion of the interface was observed, that is, length L between the lowest point a and highest point d in the illustration of Fig. 2. The extension ΔL of the interface was obtained from many photos taken by changing the applied magnetic field under fixed temperatures (65.3 or 77.1 K), and was plotted as a function of the magnetic field as shown in Fig. 3. Here, the extension ΔL of the interface is defined as

$$\Delta L = L_B - L_0 , \tag{1}$$

where L_0 and L_B are the lengths of the interface in the magnetic field of zero and B, respectively. Since deformation of the interface is accompanied by an increase in surface energy, the abscissa in Fig. 3 was scaled with the square of applied field B, considering the fact that this increase is mainly caused by the magnetic field. It should be noted that the size of the error bars in Fig. 3 is $\sim$0.5 mm for all points. It is found that ΔL increases almost linearly with an increase in the magnetic field for the magnitude of B^2 up to 4: namely, the applied magnetic field up to $\sim$2 T, under either temperature of 65.3 and 77.1 K. For higher field strengths, however, ΔL deviates from this line, gets larger, and follows a separate line. Such a deviation of the data from a line is supposed to correspond to the variation in the interfacial shape from an ellipsoidal one into a sharp-pointed one as mentioned previously.

In the meantime, Arkhipenko, Barkov, and Bashtovoi[3] have taken up phenomenological considerations regarding the fact that artificial ferromagnetic drops (or nonmagnetic drops in a magnetic fluid) deform in the magnetic field. This is applied to our case of oxygen for consideration. According to their theory, round drops of gaseous oxygen at a kinematic balance in liquid oxygen are subjected to stress caused by a large difference in the magnetization of liquid and gaseous oxygen, when the magnetic field is applied, and extended in the direction of the field until newly balanced. The force causing this deformation, that is to say, the difference of the stresses given to the gas-liquid interface of oxygen, $2\sigma/R$, is in proportion to the square of the normal component of the magnetic field with respect to the interface, and is expressed as

$$\frac{2\sigma}{R} \propto A(\Delta X^2)(H\cos\theta)^2 , \tag{2}$$

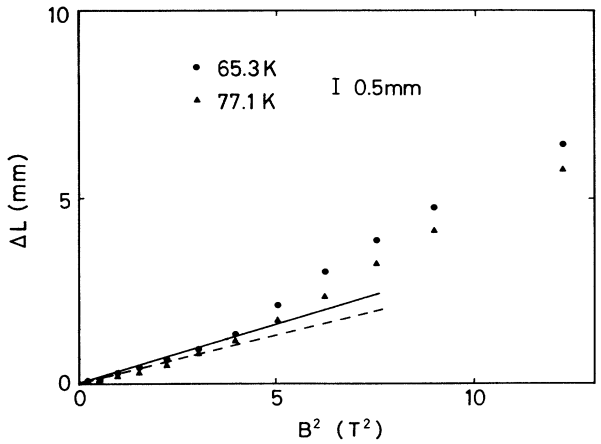

FIG. 3. Extension ΔL of the gas-liquid interface as a function of a square of the applied magnetic field B^2 at different temperatures, 65.3 and 77.1 K.

where R denotes the radius of curvature of the gas-liquid interface, σ the surface tension, θ the angle between directions of the magnetic field and normal to the interface [refer to Fig. 2(b)], ΔX the difference of magnetic susceptibilities of liquid and gaseous oxygen, and A a function mainly of $(\Delta X)^2$. In Eq. (2), however, it has been assumed that deformation of the bubble from a sphere is small and that surface tension σ does not depend on the magnetic field. First, attention is paid to points a and c_1 (or c_2) at the gas-liquid interface shown in Fig. 2. Since the angle θ becomes zero at point a and takes a certain value below 90° at point c_1, the value of the left-hand side of Eq. (2) gets larger at point a than at c_1. In other words, the radius of curvature at point a gets smaller than that at c_1 because the surface tension is the same at either point; the gas-liquid interface is considered to be lengthened in the direction of the magnetic field to suffer deformation. As already seen in Fig. 3, the relation between B^2 and ΔL, the extension of the gas-liquid interface, is approximately linear for the magnitude of B^2 smaller than 4; that is to say, the applied magnetic field below ~ 2 T, showing a trend which agrees with the contents expressed in Eq. (2). As is clear in Fig. 3, however, a magnetic field higher than this makes the shape of gas-liquid interface sharper to break the linear relationship and Eq. (2) can no longer explain the experimental results. This may be caused by the assumption of a small deformation for the sphere in the phenomenology of the magnetic fluid. Moreover, in the range of B^2 smaller than ~ 4 in Fig. 3, the extension of the interface ΔL at a temperature of 77.1 K is $\sim 20\%$ smaller than that at 65.3 K, and this is considered to be a direct reflection of the difference in the magnetic susceptibility. In fact, the calculation and comparison of A (ΔX^2) in Eq. (2) gives that for 77.1 K being $\sim 25\%$ smaller.

It is of interest to note that great interest is taken in the phenomenon in which the shape of the gas-liquid interface of oxygen gets sharp in a strong magnetic field. We consider one of the causes to be the surface tension giving a strong influence to the shape of the interface changes with the magnetic field, namely, if the surface tension has a magnetic-field effect, and we have been doing provisional experiments. As a result, it is now understood, as expected, that surface tension tends to decrease, though only a little, even in the magnetic field of ~ 1 T. This reduction in the surface tension due to the magnetic field gives rise to a decrease in the curvature radius in the left-hand side of Eq. (2), probably causing the sharpened shape of the interface.

In addition, another point of interest is the dependence of the size of the sample cell on the deformation of the gas-liquid interface in the magnetic field. As for this, research is in progress using glass capillaries with inner diameters of 1.0 and 1.5 mm. As a result of applying magnetic fields up to 3 T, it is known that the extension of the gas-liquid interface ΔL with either capillary is increased roughly in linear relation with B^2 in the temperature range from 65 to 77 K. A measurement of the surface tension and a detailed measurement of the deformation of the gas-liquid interface are in progress by increasing the magnetic field up to 7 T.

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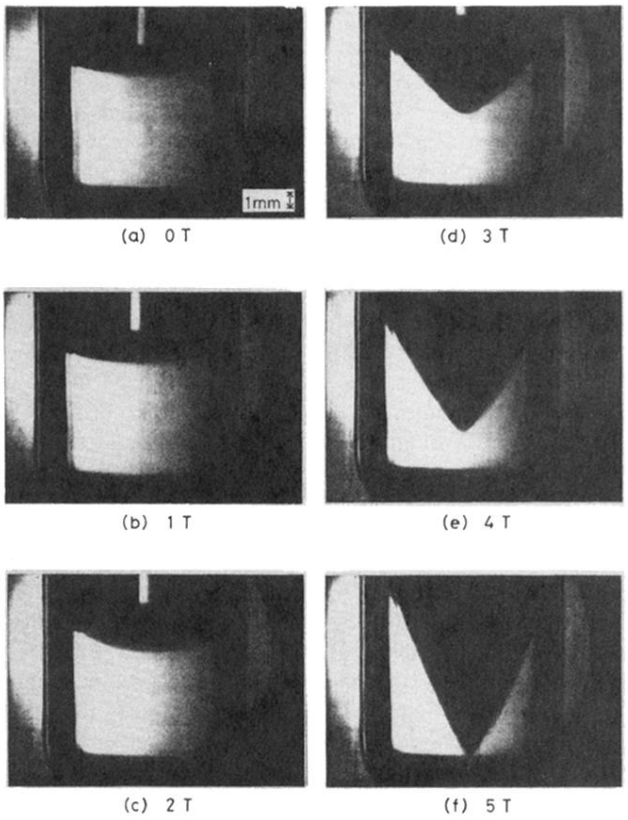

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