Surface instabilities on liquid oxygen in an inhomogeneous magnetic field

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Liquid oxygen exhibits surface instabilities when subjected to a sufficiently strong magnetic field. A vertically oriented magnetic field gradient both increases the magnetic field value at which the pattern forms and shrinks the length scale of the surface patterning. We show that these effects of the field gradient may be described in terms of an "effective gravity," which in our experiments may be varied from 1 g to 360 g.

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During the course of levitation and magnetic flotation experiments, the surface of liquid oxygen has been observed to exhibit an instability towards corrugation when subjected to a sufficiently strong perpendicular magnetic field [1]. Liquid oxygen (LOX) is strongly paramagnetic, a fact that is simply explained by Hund's rule which dictates that the two electrons in the antibonding $2\pi_g$ orbitals of the O_2 molecule form a spin triplet. This strong paramagnetism leads to a number of interesting properties of LOX when subjected to magnetic fields such as magnetic flotation [1]; the magnetovolume effect [2], in which the volume of the liquid changes under an applied field; field-induced transparency [3], in which the liquid loses its blue color.

For levitation and magnetic flotation, a large inhomogeneous magnetic field is required [4] and we observe that the presence of a vertical magnetic field gradient strongly influences both the magnitude of the fields required for the onset of this surface instability and also the characteristic length scale of the patterns observed. It is the influence of the field gradient which is the subject of this paper.

Surface instabilities were first investigated in ferrofluids [5,6] which consist of colloidal suspensions of ferromagnetic particles (such as magnetite or cobalt) in organic liquids (such as oil or kerosene) to which are added surfactants which coat the particles in order to inhibit cohesion and prevent field-induced flocculation.

If a sufficiently strong magnetic field is applied perpendicular to the surface of a layer of magnetic fluid that surface will develop static corrugations [5,6]. The appearance of these surface corrugations lowers the magnetic energy of the liquid because of the focussing of the magnetic flux toward the peaks in the surface pattern. Such corrugations, however, cost gravitational energy (in moving fluid from the troughs to the peaks) and surface free energy (by increasing the total area of the surface). Only when the gain in magnetic energy exceeds the cost in gravitational and surface free energies will the surface spontaneously corrugate. Above a lower critical field, the surface corrugations form a triangular lattice. Figure 1 shows our own observation of such a pattern on LOX. At higher fields, however, a square lattice is observed. For our own observations of this on LOX see Fig. 2.

Cowley and Rosensweig [5] carried out a linear stability analysis to determine the critical field for the onset of the instability and the wavelength of the corrugations at the onset, for the case of a ferrofluid subject to a uniform field. This work was extended by Gailitis [7,8] (see, Ref. [9], and references therein for more recent work) to include the first nonlinear corrections in order to examine the criteria for pattern selection. The theory of surface instabilities in the presence of inhomogeneous fields was developed by Zelazo and Melcher [10] and the principal effect of the field gradient can be regarded as a renormalization of the acceleration due to gravity, $g \rightarrow G = g + f_p$, where f_p is the force driving unit mass of paramagnetic fluid towards the higher field region.

In this paper we test the supposition that this is the principal effect of the field gradient. Following a brief description of our experimental arrangement we give a brief outline of the results of Zelazo and Melcher [10] as they apply to our system, along with specific predictions for the critical applied field for surface corrugation and the wavelength of the surface pattern at the onset of the instability. Finally we compare these predictions with our experimental observations testing the validity of the theory.

The experiments were conducted using a specially constructed 17-T-superconducting Bitter-solenoid magnet having a 50-mm-diameter vertical bore. For this magnet, the maximum value of $B_0 dB_0/dz$ is 1470 $\mathrm{T}^2\,\mathrm{m}^{-1}$ offering an "effective gravity" of up to 360 g for LOX. A pool of LOX 1.5 cm deep was contained in a glass Dewar vessel (see Fig. 3).

The liquid was held at its boiling point at atmospheric pressure. It was slowly lowered into the bore of the magnet, until a pattern of peaks was observed. The magnet is designed to enhance radial variations in the vertical field, so that the corrugation pattern first appears with perfect triangular symmetry in the center of the fluid surface. At higher fields the pattern covers the entire surface but the circular

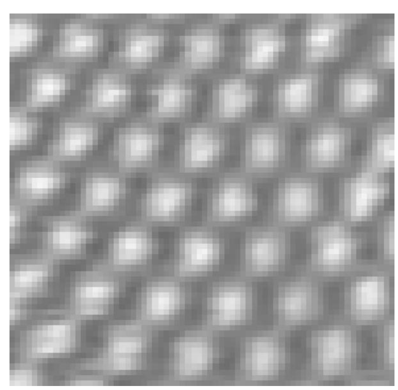

FIG. 1. View of the hexagonal patterns formed on the surface of liquid oxygen held inside a glass Dewar within the bore of the magnet. The field of view is $\sim 17 \times 17$ mm^2.

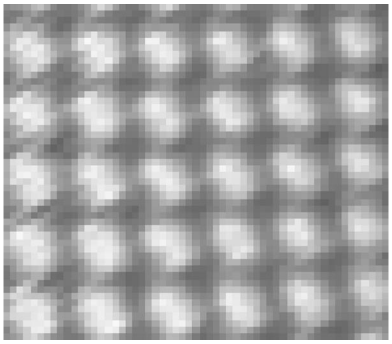

FIG. 2. View of square patterns formed on surface of LOX. The field of view is $\sim 6.5\times 5.5\ \mathrm{mm}^2$.

container walls force dislocations to appear in the regular lattice. The magnetic field and the magnetic field gradient could be adjusted by the use of various magnet currents and liquid surface positions.

A charge-coupled device camera (CCD) was used to display a magnified image of the surface of the LOX on a television screen, aiding the observation of the onset of surface instabilities and facilitating more accurate measurements of the spatial separation of the peaks.

The instability to surface corrugation only depends on the local properties of the field profile at the surface of the liquid oxygen which can be characterized by the field at the surface, B_0, and the length scale for field variations, ζ, defined by (recall that the field decreases with z, the distance from the center of the magnet)

$$\zeta^{-1}=\left(\frac{-1}{B(z)}\frac{\partial B(z)}{\partial z}\right)_{\mathrm{surface}}. \tag{1}$$

The relevant material parameters are the density ρ, the surface tension σ and the susceptibility χ of the oxygen. We

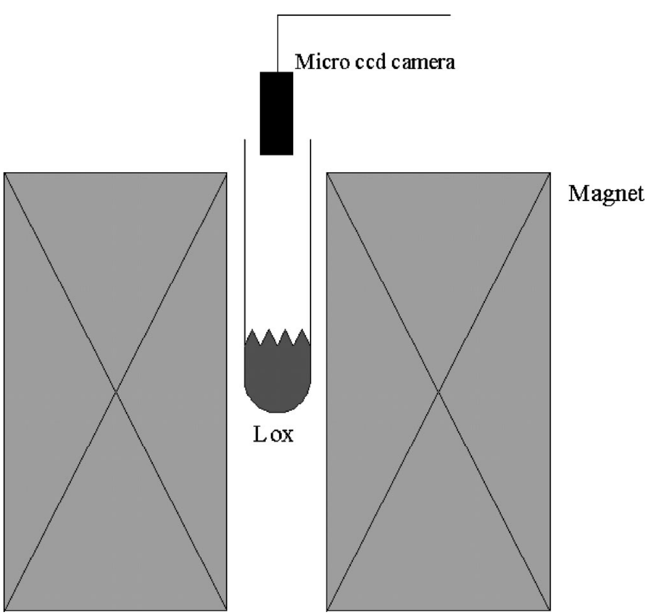

FIG. 3. Experimental setup.

take these to have the following values at 90 K: $\rho = 1149\ \mathrm{kg\,m^{-3}}$; $\sigma = 1.32\times 10^{-2}\ \mathrm{Nm^{-1}}$; and $\chi = 3.47\times 10^{-3}$ [11].

Zelazo and Melcher [10] carried out an analysis of the dynamical behavior of a ferrofluid surface subject to an inhomogeneous applied field. The coupled equations for the magnetic field and the fluid were solved to lowest order in the distortion of the surface to arrive at the dispersion relation for surface modes. The situation in our experiments is simpler in several respects than the general case considered in Ref. [10]. First, the depth of the LOX layer and the height of the air space above it are both much larger than the typical length scales for the observed corrugations so that the fluid and the air above it may be considered to occupy infinite half spaces. Second, the magnetic response of the oxygen is essentially linear over the whole range of fields that we consider, so that the difference between B/H and $\partial B/\partial H$ can be neglected. With these simplifications the prediction of Ref. [10] for the frequency of the surface mode with wave number k is given by

$$\rho\omega_k^2=\sigma k^3-\lambda\frac{B_0^2}{\mu_0}k^2+\rho G(B_0,\zeta)k, \tag{2}$$

where the coefficient

$$\lambda=\frac{\chi^2}{2(2+\chi)(1+\chi)}\sim\frac{\chi^2}{4},\quad \chi\ll 1 \tag{3}$$

accounts for the energy gain (and the hence decrease in the restoring force) due to the focusing of field lines through peaks in the surface and

$$G(B_0,\zeta)=g\left(1+\frac{\chi}{\rho g(1+\chi)\zeta}\frac{B_0^2}{\mu_0}\right) \tag{4}$$

is the effective acceleration due to gravity, including the attractive force on the paramagnetic oxygen towards the center of the magnet.

A static distortion will appear with wave number

$$k=\frac{\lambda B_0^2\pm\sqrt{\lambda^2B_0^4-4\mu_0^2\rho\sigma G(B_0,\zeta)}}{2\mu_0\sigma} \tag{5}$$

provided the argument of the square root is positive. Hence, there is a critical value of the field below which no such static distortion exists

$$B_c(\zeta)\approx\frac{8}{\chi}\sqrt{\frac{\mu_0\sigma}{\chi\zeta}},\quad \chi\ll 1. \tag{6}$$

For comparison with the experiment it is useful to define $\tilde{g}(\zeta)=G(B_c(\zeta),\zeta)$, the value of the effective gravity at the onset field for a given field gradient. The following relation should then hold

$$B_c^4=\frac{4\rho\sigma g}{\lambda^2}\left(\frac{\tilde{g}}{g}\right). \tag{7}$$

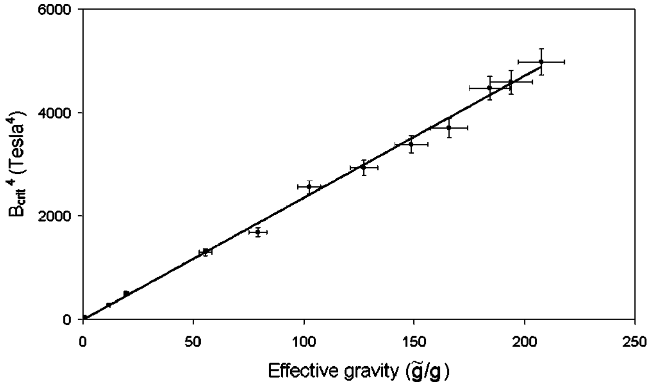

FIG. 4. The fourth power of the onset field for the hexagonal pattern, B_c, plotted against the effective gravity $\tilde{g}/g$. The bars indicate the accuracy of the data. The continuous line is a best fit to the data.

The wave number of the static mode at the critical field is then

$$k_c = \frac{\lambda B_c^2}{2\mu_0 \sigma} = \sqrt{\frac{\rho g}{\sigma}} \left(\frac{\tilde{g}(\zeta)}{g} \right)^{1/2}. \tag{8}$$

The spacing between the peaks, in the triangular lattice observed, should then be

$$L = \frac{4\pi}{\sqrt{3} k_c} = 4\pi \sqrt{\frac{\sigma}{3\rho g}} \left(\frac{\tilde{g}(\zeta)}{g} \right)^{-1/2}. \tag{9}$$

In order to test Eq. (7), in Fig. 4 we display the experimental data as a plot of B_c^4 against the effective gravity at onset, $\tilde{g}/g$. An estimate of the experimental accuracy of each datum point is shown. The data are well fitted by a straight line passing close to $\tilde{g}/g = 1$ at $B_c = 0$ and having a slope of (23.7 ± 0.8) T^4 confirming the expected dependence of B_c upon $\tilde{g}$. The dominant error arises from the determination of the point of instability. Using the values quoted above, Eq. (7) predicts

$$B_c^4 \approx \frac{\tilde{g}}{g} (26.4) \ (\mathrm{T}^4). \tag{10}$$

The discrepancy between theory and experiment is marginally significant and will be the subject for further investigations.

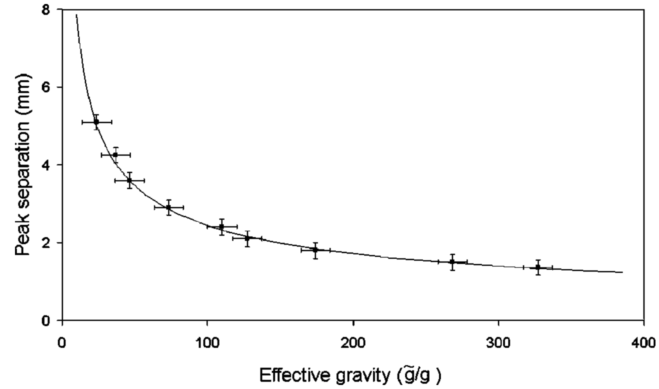

FIG. 5. The peak spacing L for fields just above onset, plotted against effective gravity $\tilde{g}/g$. The continuous line is the prediction derived from Eq. (9), using the values for ρ and σ given in the text.

The measured peak separation at onset, L, is plotted against $\tilde{g}/g$ in Fig. 5, together with the prediction of Eq. (9) that $L^2\tilde{g}/g = 6.05 \times 10^{-4}$ m^2 based on the values of ρ and σ given above. The prediction fits the data very well, a best fit to the data corresponding to $L^2\tilde{g}/g = (6.2 \pm 0.3) \times 10^{-4}$ m^2.

To summarize, we have observed the corrugation instability [5] in the surface of liquid oxygen and examined the dependence of the critical field and wavelength of the pattern on the field gradient. We have found that, in agreement with the theory developed for ferrofluids, the principal effect of a field gradient is to renormalize the gravitational acceleration in the manner described by Eq. (4), the experimental observations offering satisfactory agreement with the theoretical predictions for the onset field and the wavelength over the wide range of effective gravities which we have examined. Liquid oxygen is not only a homogeneous elemental magnetic liquid suitable for the study of fluid and lattice dynamics over a very wide range of effective gravities, but also offers a low cost environmental friendly fluid for a range of technological applications such as the separation of precious minerals.

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