Nonlinear Deformation of the Electron-Charged Surface of Liquid ^{4}He

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Deformations of the charged surface of liquid ^{4}He are studied by optical means. Near the dimple-crystal–formation point we observe saturation effects and a stripe structure exhibiting hysteresis. We discuss imperfect lattices and dimple glasses and compare data with existing theories of nonlinear equilibrium.

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The electron-charged surface of liquid ^{4}He has been the subject of numerous studies in recent years.[1] For sufficiently large electric fields (2–3 kV/cm), coupling of the two-dimensional electron gas with the fluid motion of the underlying helium results in a capillary wave instability and the ensuing "dimple crystal" first observed by Wanner and Leiderer.[2-5] In this Letter, we report measurements of the nonlinear behavior of the surface near this instability point, including saturation effects and the observation of a precursor, "stripe" structure which exhibits considerable hysteresis. We compare our data with two theories of the dimple crystal. In addition, we discuss the character of imperfect dimple crystals: grain boundaries, dimple "glasses," and the growth time for these structures.

Our experiments were carried out in a vibrationally isolated cryostat with a vertical optical axis. The liquid helium (in equilibrium with its vapor) was cooled indirectly from a pumped ^{4}He chamber. Surface deformations of the liquid were used to refract a parallel beam of incoherent light, thus imaging the space-time variations of the liquid surface curvature. The light-intensity variations were then monitored with a vidicon-tube television camera and stored with a video recorder. The recorder output voltage provided a quantitative measure of the light intensity reaching the camera. The actual helium under study was constrained in a rectangular geometry, between two electrically conducting glass plates separated by 0.528 ± 0.0005 cm across which electric fields up to 3 kV/cm could be applied. The electron source was a glow discharge. Electron density was obtained by collecting the electrons with a surrounding guard electrode and measuring the charge with an integrator.

In the deep liquid limit, the dispersion relation for the charged surface is given by[6]

$$(\rho_l+\rho_g)\omega^2 = (\rho_l-\rho_g)gk+\alpha k^3-(k^2/4\pi)(E_+^2+E_-^2), \quad (1)$$

where $E_+ - E_- = 4\pi n_s e$, with n_s the surface electron density. E_+ and E_- are the static fields above and below the surface, g the acceleration of gravity, α the surface tension, and ρ_l, ρ_g the liquid and gas densities, respectively. When the vertical electric field exceeds a critical value given by

$$\tfrac{1}{4}(E_+ + E_-)^2 = E_0^2 = 4\pi[(\rho_l-\rho_g)g\alpha]^{1/2} - (2\pi n_s e)^2, \quad (2)$$

the surface wave softens ($\omega \to 0$) and a hexagonal dimple-crystal dimple appears with wave number k_c, given by

$$k_c = [(\rho_l-\rho_g)g/\alpha]^{1/2}. \quad (3)$$

Our measurements in a temperature range 1.5–4.2 K have confirmed (2) and (3) to within an overall error of ~2%, arising from slight plate misalignment, liquid-level uncertainty, and existing data for surface tension and liquid density.[7]

To study the nonlinear behavior of surface deformations for $E > E_0$ we measure the dimple depth[8] as a function of electric field increment, $E - E_0$. We use the experimental E_0 at which a surface structure first appears since $E - E_0$ is small relative to our experimental uncertainty for the absolute critical field obtained from Eq. (2). A detail of the surface depression, a_0, is shown in Fig. 1(a). Starting at A, the initially flat surface deforms first into a shallow "stripe" structure (shown in Fig. 2), and then quickly jumps to a much larger value of a_0, at which point the dimple lattice is observable. As the field is increased the depth quickly saturates for $E - E_0 \gtrsim 10$ V/cm. The dimple radius is found to be independent of depth. Experiments at different electron densities in the range $(1-5)\times 10^7/\text{cm}^2$ show that the saturated amplitude is proportional to the charge density. Once the dimple lattice is formed, we can draw charge off the surface with the guard electrode and leave a few isolated dimples which interact via Coulomb repulsion at

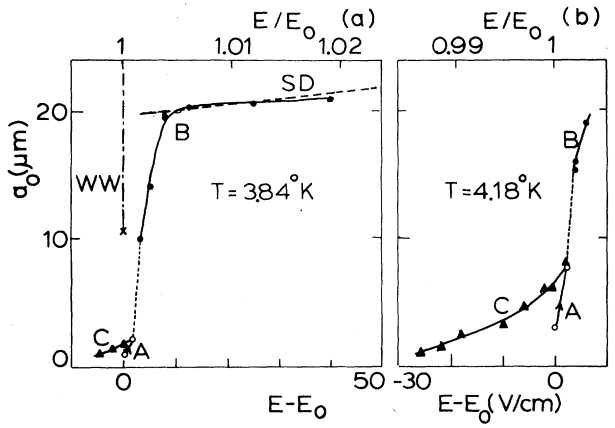

FIG. 1. (a) Depth of surface depressions, a_0, vs field increment, $E-E_0$. The segment labeled C represents hysteresis for the strip deformation. (b) Depth of surface deformation vs $E-E_0$, showing large hysteresis (segment C) for the stripe at $T = 4.18$ K.

distances $\gg 2\pi/k_c$. The saturation effect is negligibly influenced by the presence of the lattice, i.e., a single dimple exhibits essentially the same behavior as a dimple constrained in a lattice. Finally, as the electric field is increased the dimple depth rapidly increases and we lose the charge. The expanding wake produced by this "punch through" has a phase velocity characteristic of an *unchanged* surface, indicating that there are no electrons between the dimples.

In Fig. 2 (top row, left to right), as the field is increased from below critical, the appearance of stripe patterns precedes the formation of a complete dimple structure. This structure becomes clearly observable for temperatures $\gtrsim 3.4$ K, in a narrow range of electric field $|(E-E_0)/E_0| \lesssim 0.002$ below the value for the first appearance of dimples. The coexistence of the two structures near E_0 results from slight variations in field intensity and liquid depth across the cell. At the temperature shown, dimples form out of the stripe depressions and a perfect lattice is difficult to obtain. Very slow cycling of the electric field eventually produces a perfect dimple lattice. When the field is lowered below its original critical value, a surface distortion of crooked stripes persists as shown in Fig. 2 (bottom row, right to left). By repeating, several times, the cycling process shown in Fig. 2, we have observed hysteresis as large as 20 V/cm below the original critical field. In Fig. 1(b), we plot the total amplitude of the surface depression (at a fixed region in the cell) as a function of $E-E_0$. If we start from $a_0=0$ and E exceeds E_0, then the stripe amplitude increases along the line labeled A. The amplitude then jumps to the curve B once dimples are formed. After the dimple state is created, a decrease of $E-E_0$ results in crooked stripes with amplitude following the curve C. Hysteresis effects of order 10 V/cm have been reported by Ebner and Leiderer,[4] although they do not mention whether this effect is associated with stripe patterns or dimples. The amplitude of our hysteresis is largest near 4.2 K and becomes unobservable below ~3.7 K. The maximum amplitude of the stripe is also an increasing function of temperature.

We now compare our amplitude data to two existing theories for the surface deformation: a wave-wave (WW) coupling theory[9] and a single-dimple (SD) theory.[10] The wave-wave coupling theory is based on an assumption that the perturbed electron density is small relative to the unperturbed density, so that the surface is always an equipotential. The dotted line labeled WW in Fig. 1(a)

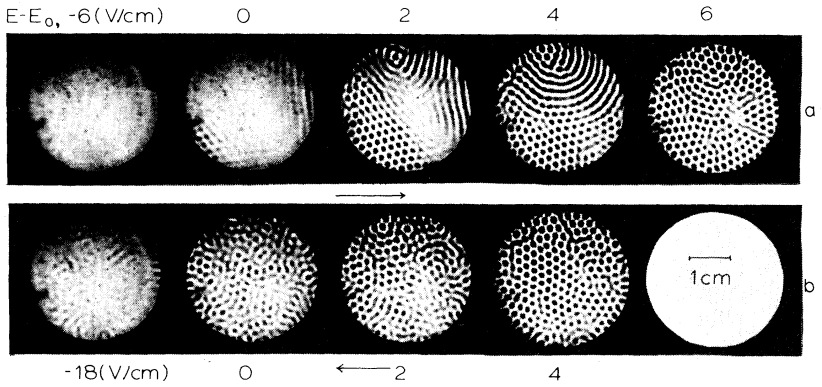

FIG. 2. Shadowgraph pictures, showing hysteresis. The field is increased in row (a) and decreased in row (b). The stripe deformation is clearest in the top row.

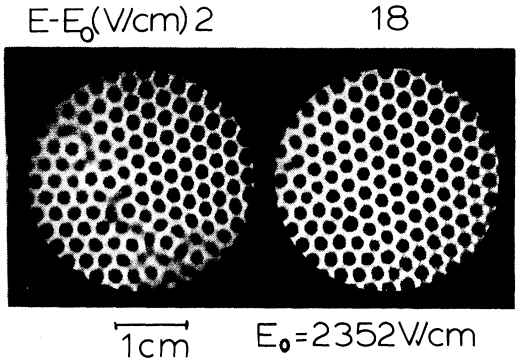

FIG. 3. Dimple-crystal grain boundaries for two different values of electric field.

is the prediction of theory for small values of $E - E_0$. The theory predicts a first-order jump in a_0, followed by a rapid increase with field increment. It also predicts a slight (~0.01 V/cm) hysteresis for the dimple lattice, but this effect is not within our experimental resolution. The theory does not account for stable stripe perturbations.

The single-dimple theory postulates that charge is localized to a small region of order $(1/k_c)$ within each dimple while the surrounding liquid surface is neutral. It predicts a first-order jump in a_0 followed by a saturated amplitude proportional to the *total* electric field and is consistent with our results, as shown by the line labeled SD in Fig. 1. The region observed between the initial jump at lattice formation and the saturation at B is not accounted for in either theory. An extension of the SD energy calculation also accounts for the stripe instability at low values of $E - E_0$. Physically, the stripe is favored at small $E - E_0$ because of the logarithmic dependence of the Coulomb repulsion energy upon stripe width. However, the temperature dependence of the stripe phenomena is not accounted for in this calculation.

In the temperature range 2.5–3.4 K, where the stripe feature is not observable, we consistently observe regions of perfect hexagonal lattice separated by grain boundaries. For small values of $E - E_0$, these boundaries appear as shallow, very slowly moving groove depressions connecting neighboring dimples. As the field is increased, the grooves disappear and the grain boundaries become fixed with characteristic pairs of dimples showing coordination numbers of 5 and 7, respectively. This behavior is illustrated in Figs. 3(a) and 3(b). The grain boundary angle is typically ~30°.

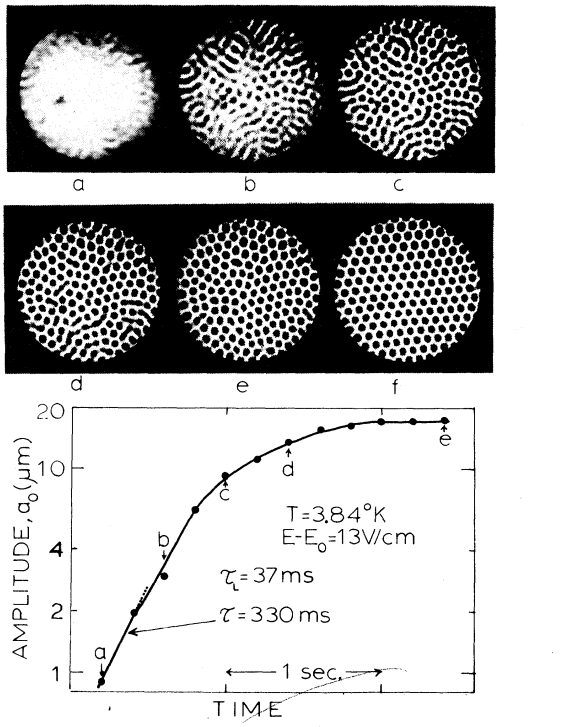

FIG. 4. Time development of the surface deformation after a sudden field change. A glassy structure appears (e) but a perfect crystal (f) can eventually be formed with slow cycling of the electric field. τ_L is the linear growth rate, given by $[2\pi\alpha/g(E^2 - E_0^2)]^{1/2} = \tau_L^{-1}$.

Once the stripe feature becomes observable ($T \gtrsim 3.4$ K), the formation of the perfect lattice is hindered by a propensity towards glass formation, as mentioned earlier, and illustrated in Fig. 2. In Fig. 4, we demonstrate this behavior by "quenching" the charged surface, i.e., abruptly stepping the electric field from $E < E_0$ to $E - E_0 = 13$ V/cm. The real time evolution of the surface is shown in Figs. 4(a)–4(e). At this temperature, the time constant for complete saturation of surface amplitude is ~330 msec, a full order of magnitude higher than the growth time predicted by linear instability theory. The inclusion into the charged-surface dispersion relation of an electronic mobility that is finite, rather than infinite, still yields a growth time that is too small to account for the long growth times observed. As yet, we do not understand this feature of the experiments.

In conclusion, we find that the surface amplitude behavior favors a localized charge model. We observe a stripe deformation whose tempera-

ture-dependent hysteresis is not understood. While the physics of this system is essentially determined by fluid mechanics and electrostatics, the nonlinearity of the problem also results in many phenomena relevant to microscopic two-dimensional electron systems.

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$$I(\vec{r}+\gamma\nabla a)/I_0=[1+\gamma(a_{xx}+a_{yy})+\gamma^2(a_{xx}a_{yy}-a_{xy}^2)]^{-1},$$

$\gamma=l/(n-1)$, where I_0 is the intensity when $a=0$, l is the distance from the liquid surface to the screen, n is the index of refraction of the liquid helium, and the subscripts x and y stand for the partial derivatives with respect to those quantities. The derivatives of a must be evaluated at $\vec{r}$ $(\equiv x\hat{x}+y\hat{y})$.
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Interaction of Hydrogen Atoms with Helium Films: Sticking Probabilities for H on ^{3}He and ^{4}He, and the Binding Energy of H on ^{3}He

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Magnetic resonance at 1420 MHz in zero magnetic field and for $0.06<T<0.5$ K has been used to determine the binding energy of H on liquid ^{3}He, the rate constant for recombination and the frequency shift for H on ^{3}He, and the sticking probability for H on ^{3}He and ^{4}He. The binding energy for H on liquid ^{3}He is found to be 0.42 ± 0.05 K, and the sticking probabilities are 0.035 for H on ^{4}He and 0.016 for H on ^{3}He.

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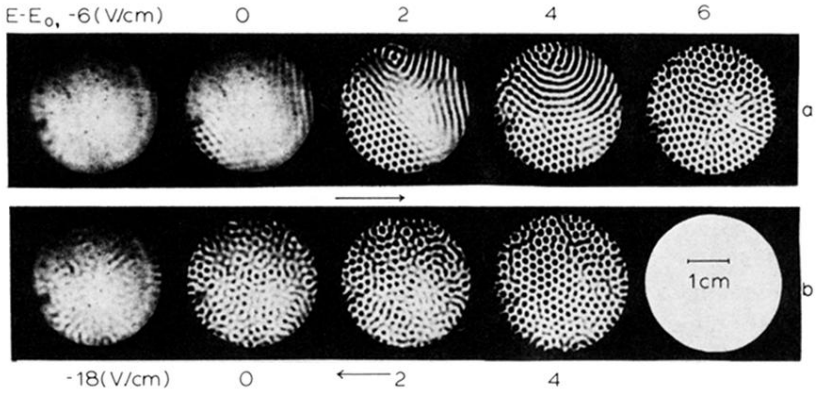
Because hydrogen atoms bind strongly to any solid surface, all low-temperature experiments on atomic hydrogen gas[1-6] must be performed in containers which are coated with a liquid-helium film. Hydrogen also sticks to liquid helium, but the binding energy is much smaller than for solids. Earlier magnetic-resonance studies in our laboratory[4] have shown that the binding energy E_B for H on liquid ^{4}He is 1.15 ± 0.05 K. A similar but somewhat lower result, $E_B=0.89\pm0.07$ K, has been reported by Matthey, Walraven, and Silvera.[5] In this Letter we show that the binding energy of H on liquid ^{3}He is about 0.4 K in agreement with very recent results of van Yperen *et al.*[6] at high magnetic field. It would appear that this value represents the minimum possible binding energy for H atoms on the walls of a container.

The minimum value of E_B has important consequences for experimental attempts to observe Bose-Einstein condensation in a high-density gas of spin-polarized H. Because the surface state lies below the continuum, there is a strong tendency at low temperature for H atoms to reside on the surface. The surface density is eventually limited by repulsive interactions between H atoms, and only then can the addition of more atoms lead to a macroscopic occupation of the lowest-energy bulk states of the gas. The saturated surface density n_w^s is proportional to E_B, $n_w^s=\gamma E_B$, where[7,8] $\gamma=(0.5-1.0)\times10^{14}$ K^{-1} cm^{-2}. The main obstacle to the achievement of Bose-Einstein condensation then lies in the fact that such high surface densities are unstable against the recombination process $H+H\rightarrow H_2$ on the wall.

We also report measurements of the wall recombination cross section λ for H on ^{3}He in zero magnetic field which we find to be nearly equal to that for H on ^{4}He. Direct comparison to the high-



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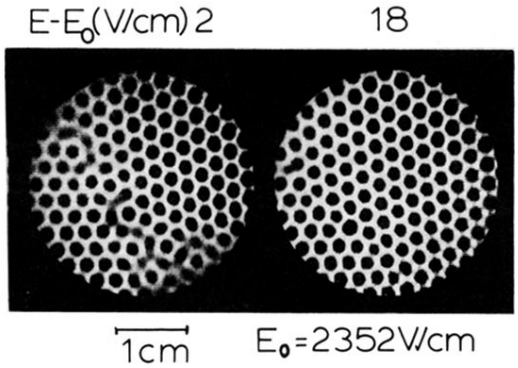

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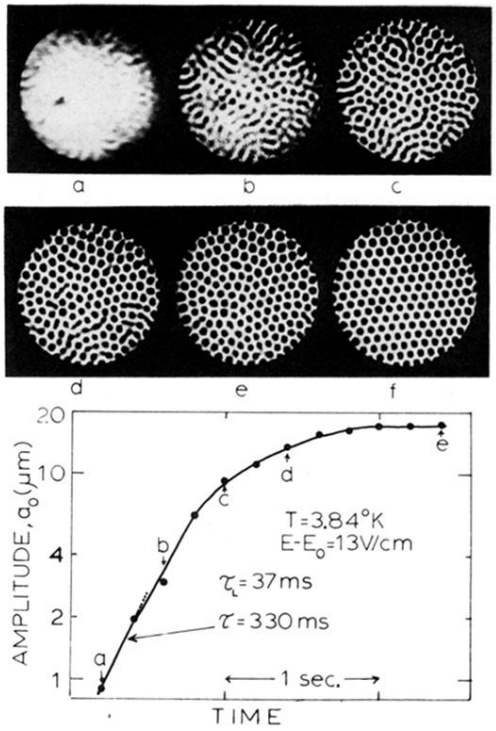

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