Charged interface of ^{3}He-^{4}He mixtures. Softening of interfacial waves

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The excitation spectrum of ripplons at the charged interface of phase-separated ^{3}He-^{4}He mixtures has been determined for electric fields E up to E_c, given by the stability limit of the interface. The frequency $\omega/2\pi$ of the ripplons is substantially reduced by the presence of the charges, most pronounced for excitations with a wave vector q_c equal to the inverse capillary length, where $\omega \to 0$ when the electric field approaches the critical value E_c. Both E_c and q_c depend on temperature via the interfacial properties. In the investigated temperature region, $0.5 < T < 0.77$ K, and for two types of charge carriers, negative and postitive ions, the experimentally determined ripplon softening is well accounted for by the theory.

I. INTRODUCTION

The surface of a liquid is normally subject to two forces due to gravity and surface tension. Both counteract deviations of the surface from its equilibrium position, and are thus responsible for the stability of the liquid surface.

When an external electric field is applied perpendicular to the surface, an additional force appears which, as will be shown further below, reduces the stabilizing effect of gravity and surface tension. As a result, the spectrum of excitations of the surface is altered markedly, and for high enough fields even a (so-called electrohydrodynamic) instability of the surface is expected.

For a well-conducting liquid, like a liquid metal, the influence of the electric field has been studied theoretically for the first time by Frenkel.[1] An isolating liquid should behave similarly, except that larger fields are necessary to reach the instability threshold.[2,3] The following considerations not only apply to a usual liquid surface — which strictly speaking is an interface between coexisting liquid and gas — but also to the interface between two stratified liquids.

Since the external electric field reduces the restoring forces at the surface, it is evident that the frequency of surface waves — also called ripplons — will be lowered as the field is raised. This "ripplon softening" should be most pronounced for a particular wave vector q_c. At a critical field E_c the frequency of ripplons with $q = q_c$ is predicted to vanish, which implies that then elongations of the surface are no longer driven back — the surface becomes unstable.

Despite this expected prominent effect experimental results on that subject are rather sparse. Field-induced breakdown at the interface of liquid insulators has been investigated, e.g., by Taylor and McEwan,[4] and for the special case of a thick He film by Cole *et al.*[5] A decrease of the frequency $\omega/2\pi$ of surface excitations (at a few fixed q values) was observed by Melcher,[6] but the experimentally accessible region was so small that the reduction in ω amounted to only a few percent. Another interesting system where an instability was found recently is the free surface of liquid He charged with electrons.[7]

We would like to present here the most detailed results for the development of an electrohydrodynamic instability to date. The system we have chosen is the charged interface of phase-separated ^{3}He-^{4}He mixtures. Particularly interesting in this system is the vicinity of the tricritical point (temperature $T_t = 0.867$ K, ^{3}He concentration $x_3 = 0.675$), where the properties of the interface, like the interfacial tension, vary strongly with temperature. Since the conditions for the instability depend crucially on the interfacial parameters, these measurements can provide a thorough test of the theory.

As has been shown in an earlier experiment, it is possible to hold charge carriers at the ^{3}He-^{4}He interface for a considerable length of time provided the temperature is not too close to T_t.[8] These charges, which are positive or negative ions ("snowballs" and "electron bubbles," respectively[9]), then form a two-dimensional Coulomb system at the interface. If the interface is charged to saturation, so that in one of the two coexisting helium phases the externally applied electric field is screened completely by the ions, this system corresponds to a liquid conductor. Partially charged interfaces were investigated, too, and they were found to exhibit a new instability behavior, distinctly different from the chaotic breakdown observed as a result of the electrohydrodynamic instabilities studied until now.

In this first part we describe the properties of the interface-ion system for electric fields smaller than the critical field E_c, i.e., that region where softening of the ripplons should be observable. In a second part[10] we shall show what happens for fields $E > E_c$, where the instability of the interface develops. A brief report of this work has already been published.[11]

II. INTERFACIAL EXCITATION SPECTRUM

A. Uncharged interface

The motion of a normal liquid interface, at rest located in the x-y plane, is governed by[12]

$$(\rho_- + \rho_+)\frac{\partial \Phi}{\partial t} = (\rho_- - \rho_+) g \xi + \sigma \frac{\partial^2 \xi}{\partial x^2} \quad . \tag{1}$$

Here ρ_- and ρ_+ are the densities of the lower and the upper phase, respectively, Φ is the velocity potential, g is the acceleration of gravity, σ is the interfacial tension, and ξ is the vertical elongation of the interface from its equilibrium position. In Eq. (1) we have assumed that ξ is varying only in the x direction.

Setting $\xi = \xi_0 e^{i(qx-\omega t)}$ and bearing in mind that $\partial\Phi/\partial z = \partial\xi/\partial t$, we obtain the dispersion relation for interfacial waves of frequency $\omega/2\pi$ and wave vector q

$$(\rho_- + \rho_+)\omega^2 = (\rho_- - \rho_+) gq + \sigma q^3 \quad , \tag{2}$$

where we have made the simplifying assumption, always fulfilled in our experiment, that the distances d_+ and d_- of the interface from the container top and bottom are large enough that $\tanh(d_+ q) \simeq \tanh(d_- q) \simeq 1$.

For small q the first term on the right-hand side of Eq. (2) dominates, yielding $\omega \propto q^{1/2}$ for these "gravity waves"; for large q the dispersion of "capillary waves", $\omega \propto q^{3/2}$, is obtained. The transition between these two types of waves occurs around $q = 1/a$ where

$$a = [\sigma/(\rho_- - \rho_+) g]^{1/2} \quad , \tag{3}$$

is the capillary length, describing the typical range of static deformations of the interface. Incidentally, the phase velocity v_{ph} of interfacial waves has a minimum at $q = 1/a$; for a charged interface the importance of waves with this particular wave vector will become more evident further below.

It was found earlier[13] that Eq. (2) yields a reasonable description also for the unique interface of ^{3}He-^{4}He mixtures, where a superfluid — the ^{4}He-rich phase — coexists with a normal ^{3}He-rich phase. The superfluidity of the lower phase apparently has no influence on the excitation spectrum, because for the interfacial waves considered here the superfluid and normal component of the ^{4}He-rich liquid are moving in phase as they do in bulk superfluid helium in a wave of first sound.[14]

So far damping of the ripplons due to the viscosities of the bulk liquid has been neglected. A more complete treatment shows that damping modifies Eq. (2), leading to a somewhat complicated implicit equation for the dispersion relation.[13,15] Since in the region of interest the resulting correction of the ripplon frequencies is only of the order of 5%,[16] we omit here damping for the sake of simplicity.

B. Charged interface

It is known that sufficiently far below the tricritical point the interface of ^{3}He-^{4}He mixtures presents a barrier at which ions can be bound.[8] At temperatures around 0.5 K the direction in which the barrier is effective depends on the sign of the charges: positive ions cannot penetrate from the lower into the upper phase and thus, when an appropriate electric field is applied, form a layer located about 100 Å *below* the interface. Conversely, negative ions can be accumulated in a sheet of charges just *above* the interface.

When the tricritical point is approached, the energy barrier of height ΔW holding the ions in place decreases. As ΔW becomes comparable to the thermal energy, the ions can by thermal activation cross the interface after a trapping time τ which drops exponentially for $T \to T_t$. Whereas this behavior is in principle understood, a detailed explanation is still lacking, especially for negative ions, where the current "bubble" model predicts an energy barrier distinctly smaller than the value determined from experiment. Besides, present theory can only qualitatively account for the electric field dependence of τ. In the following, however, these details of the ion trapping are not important. We restrict ourselves here to temperatures $T < 0.77$ K, where the trapping times are long enough for the ions to arrange themselves such that they form a two-dimensional layer of charges in equilibrium. The sign of the ions should be irrelevant for the effects to be studied, and, as will be seen, this is indeed the case.

When the interface is charged the ions under the influence of the (homogeneous) external electric field exert a pressure upon the interface. For a completely uniform charge distribution this pressure also is uniform. Any local elongation of the interface, however, results in a rearrangement of the ions, which are accumulated in the troughs and depleted in the crests of a, for example, sinusoidal perturbation. Therefore, the local pressure in the troughs is higher and in the crests is lower than average. The ions thus tend to increase any deformation of the interface.

Consequently in the equation of motion of such a charged interface an additional term appears due to the pressure arising from the discontinuity in the electric field perpendicular to the interface

$$(\rho_+ + \rho_-)\frac{\partial \Phi}{\partial t} = (\rho_- - \rho_+) g \xi + \sigma \frac{\partial^2 \xi}{\partial x^2} - \frac{[E(x)^+]^2 - [E(x)^-]^2}{8\pi} \quad , \tag{4}$$

where the (+) and (−) signs again refer to the upper and lower phase. We shall assume in the following that the interface is charged to the saturation density n_s; so that the average field in either the upper or the

lower phase (depending on the type of ions used) is screened completely, and in the other is

$$E_0 = 4\pi n_s e \tag{5}$$

(e is the elementary charge).

In order to determine now the modulation of the electrostatic pressure at the interface resulting from the redistribution of the ions in the presence of ripplons, we consider the electrostatic potential in the vicinity of the interface for which we set[17]

$$\phi(z=\xi) = \phi_0 + \phi_1 e^{i(qx-\omega t)} e^{-qz} \ . \tag{6}$$

ϕ_0 is the potential at the undisturbed interface and ϕ_1 in general is complex because the charge carriers are moving with a phase lag with respect to the interfacial wave as a result of their finite mobility μ. (In treatments of the motion of electrons at the free-helium surface the mobility has always been assumed to be large enough that this phase lag can be neglected.[18,19] For ions at the ^{3}He-^{4}He interface, where the mobility is many orders of magnitude smaller, this approximation is not necessarily justified.)

Using the equation of continuity for the charges moving in the electric field $E_{\parallel}$ along the interface

$$\frac{\partial n}{\partial t} = -\operatorname{div}(n\mu E_{\parallel}) = -\operatorname{div}\left[n\mu\left(\frac{\partial\phi}{\partial x}\right)_{z=\xi}\right] \tag{7}$$

and taking into account that

$$n = \frac{1}{4\pi e}\left(\frac{\partial\phi}{\partial z}\right)_{z=\xi} ,$$

we obtain (neglecting higher-order terms)

$$\phi_1 = \frac{(4\pi n_s)^2 e\mu q\,\xi_0}{4\pi n_s \mu q - i\omega} \ . \tag{8}$$

The pressure arising from the electric field is then

$$p(x) = \frac{[E(x)]^2}{8\pi} = \frac{1}{8\pi}[(4\pi n_s e)^2 + 8\pi n_s e\phi_1 e^{i(qx-\omega t)}] \ , \tag{9}$$

which, inserted in Eq. (4), leads to the dispersion relation for a completely charged interface[20]

$$(\rho_- + \rho_+)\omega^2 = (\rho_- - \rho_+)gq + \sigma q^3 - \frac{16\pi^2(n_s e)^3\mu q}{4\pi n_s e\mu q - i\omega}q^2 \ . \tag{10}$$

This relation shows that in general the ions, in addition to reducing the ripplon frequency, also add to the damping of the ripplons. The two limiting cases of Eq. (10) are:

(i) $\omega << 4\pi n_s e\mu q$ (or equivalently $v_{\rm ph} = \omega/q << E\mu$). If this condition is met Eq. (10) reduces to

$$(\rho_- + \rho_+)\omega^2 = (\rho_- - \rho_+)gq + \sigma q^3 - 4\pi(n_s e)^2 q^2 \ , \tag{11}$$

which agrees with earlier results of Gor'kov and Chernikova[18] and Mima *et al.*[19]

(ii) $\omega >> 4\pi n_s e\mu q$. In that case the last term in Eq. (10) vanishes because the mobility of the charge carriers is so small that they cannot follow the interfacial wave. The excitation spectrum is then equal to that of an uncharged interface.

From Eq. (10) it follows that the already mentioned instability of the interface, related to a vanishing ripplon frequency $\omega(q_c) \to 0$, appears at a critical charge density $n_c e = [(\rho_- - \rho_+)g\sigma/4\pi^2]^{1/4}$, corresponding to a critical field

$$E_c = [64\pi^2(\rho_- - \rho_+)g\sigma]^{1/4} \ . \tag{12}$$

The wave vector where this instability develops is $q_c = [(\rho_- - \rho_+)g/\sigma]^{1/2}$, equal to the inverse capillary length defined in Eq. (3).

III. EXPERIMENT

The measurements were made in a vibration-isolated optical ^{3}He cryostat. Figure 1 shows a sketch of the sample cell, which was similar to the one used earlier to investigate the trapping of ions at the ^{3}He-^{4}He interface.[8] Negative and positive ions could be studied alternatively. They were generated by a field emission tip or a radioactive source, and then by the externally applied field were drawn towards the interface, where they formed the two-dimensional Coulomb system.

Generation of ripplons on the charged interface was achieved with a horizontal 20-μm-thick wire

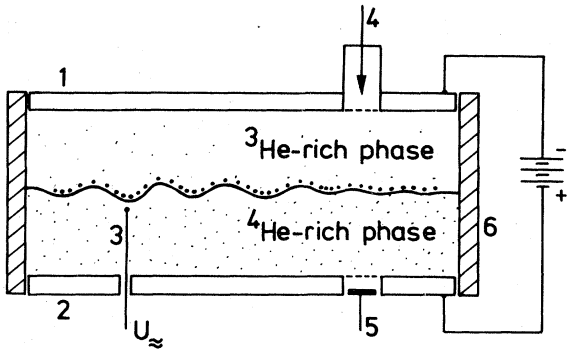

FIG. 1. Schematic sample cell for measuring the excitation spectrum of a charged ^{3}He-^{4}He mixture interface. The cell has a rectangular cross section, 20×30 mm^2, and is 5.4 mm in height. 1 and 2 are the top and bottom capacitor plates. The wire of the wave generator (3) is perpendicular to the plane of the paper, as is the plane of incidence of the scanned laser beam. In the example drawn the interface is charged with negative ions from a field emission tip (4); for positive ions a radioactive source (5) is used. The guard ring (6) is charged such that the ions cannot escape to the walls.

mounted slightly below the interface. The electric dc potential of this wire was set such that the electric holding field and therefore the ion distribution were not perturbed by the presence of the wire. The adjustment of the potential could be controlled conveniently because deviations from a uniform ion distribution lead to an inhomogeneous deformation of the interface, easily detectable by optical means.

When a small ac voltage $U_{\approx}$ (frequency $\omega_{ac}/2\pi$) is now superimposed on the dc wire voltage, then the pressure of the ions acting on the interface is modulated in the vicinity of the wire, yielding linear interfacial waves of frequency $\omega_{ac}/2\pi$. The shortest waves that can effectively be generated with such a device are of the order of the distance between wire and interface, about 0.1 mm. Since here we are mainly interested in wavelengths of the order of $2\pi a \simeq 1$ mm, this method is very well suited: a modulation of the wire potential by 1 V_{pp} is already sufficient to excite waves with an amplitude of 10^{-3} mm, a value typical for most experimental runs.

The waves generated in such a way were detected using the beam of a He-Ne laser (attenuated to 10^{-4} W), which was reflected from the interface at a small angle. The plane of incidence was parallel to the wave front of the interfacial waves, and the laser beam was focused to a small spot 30 μm in diameter, an order of magnitude less than the wavelengths to be measured. As the interfacial wave passes the detection area, the outcoming laser beam is deflected periodically by a small angle. The modulation amplitude of the deflection angle is approximately proportional to the wave amplitude at the focal area, whereas the phase of this modulation is equal to the phase of the wave at that point. Thus both amplitude and phase of the ripplons can be determined locally.

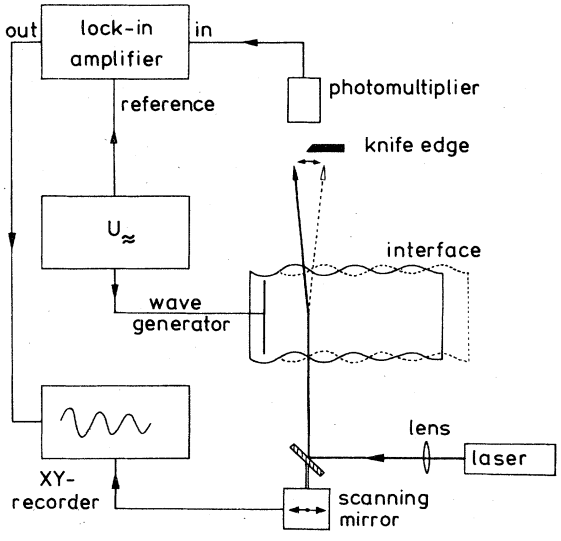

FIG. 2. Setup for measuring the ripplon dispersion relation.

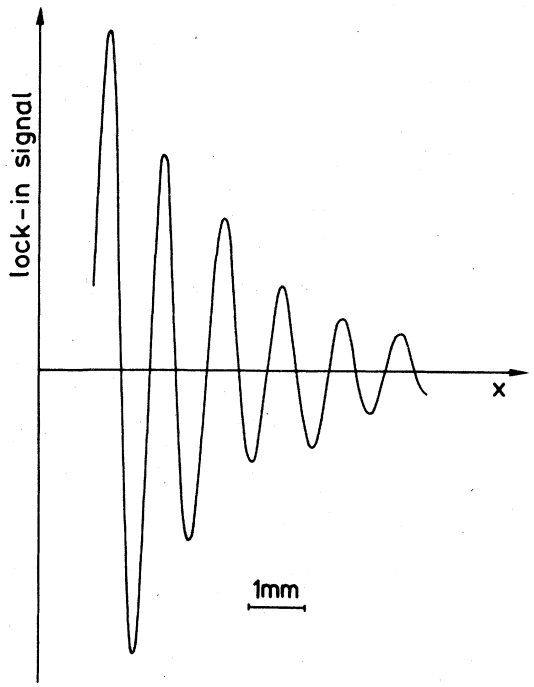

FIG. 3. Example for the output signal of the lockin amplifier as the distance x between the wave generator and the interfacial area scanned by the laser beam is varied. The pattern represents an instantaneous image of the interfacial wave. The maximum wave amplitude is about 5 μm.

A schematic drawing of the setup is shown in Fig. 2. The modulation of the reflected beam position is picked up by a photo cell located behind a knife edge. Its output is fed into a lockin amplifier which compares the phase of this signal with a reference signal from the ac voltage applied to the wave generator. When the laser spot is now scanned across the interface, a pattern is obtained which represents a momentary picture of the interfacial wave (see Fig. 3), and provides both the wavelength and the damping of that wave.

IV. RESULTS

The dispersion relation of ripplons was measured in the temperature range $0.5 < T < 0.77$ K. An example for a completely charged interface at $T = 0.67$ K, corresponding to $T_t - T = 200$ mK, is given in Fig. 4. Positive ions were used in this run.

The full line represents the dipersion of an uncharged interface as determined in an earlier experiment.[13] For low fields and correspondingly small charge densities at the interface the excitation spectrum is close to this curve. When the field is in-

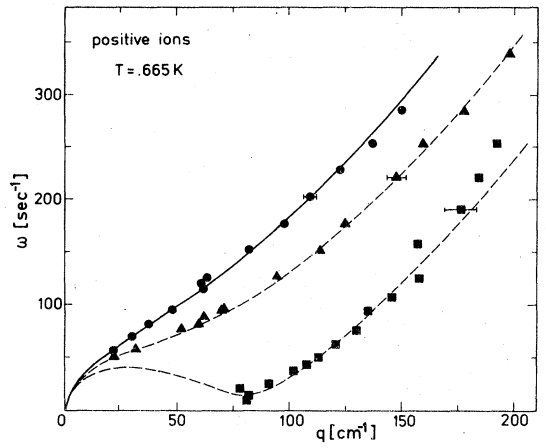

FIG. 4. Dispersion relation of ripplons at the interface of a phase separated ^{3}He-^{4}He mixture, completely charged with positive ions from below, at $T = 0.665$ K. The interface becomes unstable at $E_c = 875$ V/cm, corresponding to an ion density $n_c = 4.8 \times 10^8$ cm^{-2}. ●, $E/E_c = 0.12$; ▲, 0.71; ■, 0.995. The dashed curves are calculated according to Eq. (11). The dispersion of the uncharged interface is given by the solid line (Ref. 13).

creased, the ripplons are reduced in frequency, as is obvious in Fig. 4 for the data at $E/E_c = 0.71$.[21] This reduction becomes even more pronounced, as the field approaches the critical value E_c, which at that temperature is 875 V/cm. Apparently the largest influence on the dispersion occurs for wave vectors of the order of $1/a = 82$ cm^{-1}.

The dashed lines in Fig. 4 indicate the behavior expected according to Eq. (11). Damping of the waves due to the viscosity of the coexisting bulk phases is not taken into account here. What might appear as a more problematic assumption is that effects from the finite mobility of the ions are neglected in Eq. (11) also. The good agreement between this simplified model and the data suggests, on the other hand, that the mobility μ of positive ions along the interface is high enough to follow deformations of the interface "instantaneously". Concerning the approximation implied by Eq. (11) we can conclude, therefore, that

$$\mu >> \frac{\omega}{qE} \sim 0.005 \text{ cm}^2/\text{V sec} \ .$$

This inequality does not seem to be very informative, because it is known that positive ions have a mobility $\mu_{\text{bulk}} = 0.13$ cm^2/V sec in the bulk superfluid phase at that temperature.[22] It is not *a priori* clear, however, that the mobility parallel to the interface should not be substantially lower because of microscopic indentations that might form at the interface around the ions,[23] thus obstructing the ionic motion.

The theoretical excitation spectrum (11) displays a remarkable feature: For $E = (\frac{3}{4})^{1/4} E_c$ it has an inflection point with a horizontal tangent, and for larger values of E a region is found where three different q vectors exist for each frequency. Since in the measurement the frequency is the quantity that is fixed externally, it is not clear offhand whether only one or possibly more q vectors will be excited simultaneously. Experimentally, it was found that the wave pattern in that range was actually different from the usual exponential decay displayed in Fig. 3. The mode with the highest q appeared most clearly, but an additional modulation was present which might be interpreted as the mode of the branch with negative group velocity. An unambiguous identification turned out to be difficult because damping of the waves was considerably increased in that region. For accurate measurements of waves below $q = 20$ cm^{-1}, where the group velocity $\partial\omega/\partial q$ becomes positive again, the sample chamber was not big enough.

The parameters characterizing the ^{3}He-^{4}He interface can easily be varied by changing the temperature of the mixture. Between $T_t - T = 100$ and 300 mK the interfacial tension σ rises by a factor of 10 and the difference in density between the two bulk phases, $\Delta\rho$, by a factor of 2.5. This allows an additional check of the theory because σ and $\Delta\rho$ determine the critical field and also the critical wave vector [see Eqs. (3) and (12)]. Accordingly it is expected that the excitation spectrum should be shifted towards smaller q when $T_t - T$ is increased. Indeed, data for a temperature $T_t - T = 300$ mK, plotted in Fig. 5, show such a shift and again confirm the theoretical spectrum calculated from Eq. (11). A

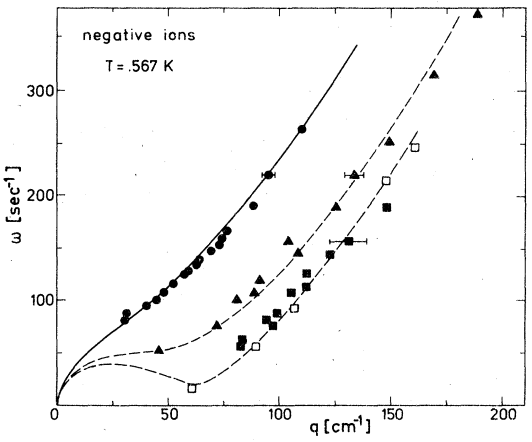

FIG. 5. Dispersion relation of ripplons at the interface of a phase separated ^{3}He-^{4}He mixture, completely charged with negative ions from above, at $T = 0.567$ K. $E_c = 1130$ V/cm. ●, $E/E_c = 0.10$; ▲, 0.88; □, 0.99. The open squares show data for positive ions at $E/E_c = 0.99$ for comparison. Again, the dashed curves are calculated according to Eq. (11), and the solid curve represents the dispersion of the uncharged interface at that temperature.

similar agreement was found to hold in the entire temperature interval investigated.

Most data of Fig. 5 were obtained with the interface charged with *negative* ions. In contrast to the positive "snowballs" these electron bubbles are located above the interface in the normal, ^{3}He-rich phase, where the bulk mobility is only $\mu_{bulk} \sim 0.03$ cm^2/Vsec.[24] Parallel to the interface the mobility might be even more reduced due to the ion-interface interaction, as argued above, but apparently also here μ is still sufficiently large not to directly affect the dipersion relation.

Of the ripplons investigated, those with the critical wave vector $q_c = 1/a$ are of particular interest, since they should finally lead to an instability of the interface at $E = E_c$. We therefore have studied waves at q_c in more detail. As the electric field is increased, the ripplon frequency should be reduced, according to Eq. (11), to

$$\omega(q = q_c) = \omega_0(q = q_c)[1 - (E/E_c)^2]^{1/2} , \quad (13)$$

where ω_0 is the frequency of ripplons at the uncharged interface. Experimental results for the ripplon frequencies at the critical wave vector for various electric fields are given in Fig. 6. Again, the dashed line shows the expected behavior, with damping not included. The datum point at E_c was obtained from a different experiment without external excitation of the ripplons, namely, from the spontaneous appearance of the spatially periodic instability with wave vector q_c.[11]

Figure 6 bears a strong resemblance to the softening of phonons, as it is observed, for example, for structural phase transitions.[25] The continuous variation of ω, with $\omega \to 0$ for $E \to E_c$, suggests that this instability can be regarded as a type of second-order phase transition, although with parameters different from the familiar quantities like the temperature. The character of this transition is not determined with high precision yet, because measurements in the immediate vicinity of E_c so far are not very accurate. Therefore it cannot be excluded with certainty that very close to E_c a finite discontinuity in ω will occur. Even that, however, would not be too unusual from the viewpoint of structural phase transitions.

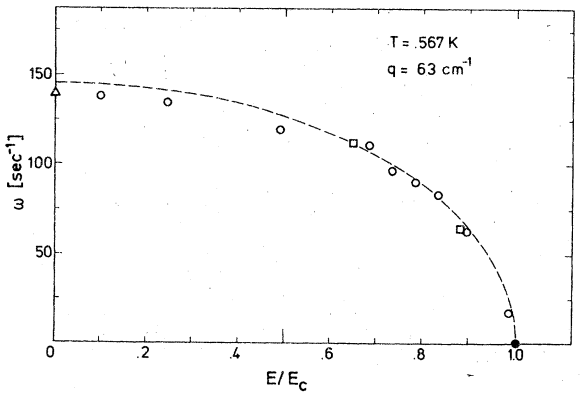

FIG. 6. Frequency of interfacial waves in ^{3}He-^{4}He at a temperature $T = 0.567$ K and a wave vector $q_c = 1/a = 63$ cm^{-1}. The interface is always charged to saturation. Circles represent measurements with positive and squares those with negative ions. The dashed line shows the result of Eq. (13). The discrepancy between this curve and the datum point at $E = 0$ (uncharged interface) is due to the damping of the interfacial waves (see Ref. 13).

V. CONCLUSIONS

The measurements reported here present the first demonstration for softening of interfacial waves over a wide range of frequencies. The specific properties of the ^{3}He-^{4}He system used in the experiments not only allowed to determine the dispersion relation of ripplons in the presence of charges, but also to vary essential parameters of the interface, and thus made a stringent test of the theory possible. Our results show that gravity-capillaray-wave excitation spectra of charged liquid surfaces indeed display the pronounced dependence on the electric field that has been predicted.

The observed softening of the ripplons is equivalent to a remarkable reduction of the phase velocity of these waves. By charging the interface this velocity could be lowered by as much as a factor of 10, e.g., from 2 to 0.2 cm/sec in the run shown in Fig. 6. As has been pointed out by Mima and Ikezi,[26] such a system with a readily variable surface wave velocity might be quite useful to study the decay of solitary waves.

The present investigations were centered around wave vectors where the changes in the excitation spectrum were expected to be most dramatic. The question arises as to what will happen at much larger wave vectors. Since the phase velocity increases as $v_{ph} \propto q^{1/2}$, a point will eventually be reached where the ions have a mobility too low to keep pace with the ripplons. Then a transition to the spectrum *without* charges should be observed [cf. Eq. (10)]. Besides, for $q > 10^5$ cm^{-1}, the average distance of the ions becomes comparable to or larger than the ripplon wavelength. In this case other modifications of the ripplon spectrum might appear, in particular when the ions are regularly distributed in the form of a two-dimensional Wigner crystal.[27]

Finally, the problem remains as to how the excitation spectrum is changed when the external electric field is increased beyond the critical value E_c, and how the interfacial instability develops. This will be the subject of a forthcoming paper.[10]

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