with Dr. K. Bedell, Dr. J. P. Blaizot, Dr. G. E. Brown, Dr. J. W. Clark, Dr. D. Coffey, Dr. S. Fantoni, Dr. V. R. Pandharipande, Dr. C. J. Pethick, Dr. D. Pines, Dr. D. G. Sandler, Dr. R. A. Smith, and Dr. Q. Usmani. The research was supported in part by National Science Foundation Grants. No. PHY 81-21399 and No. DMR 81-17182. One of us (E.K.) was the recipient of a Heisenberg Fellowship from the Deutsche Forschungsgemeinschaft.

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[1] J. Wilks, *The Properties of Liquid and Solid Helium* (Clarendon, Oxford, 1967).

[2] J. C. Wheatley, Rev. Mod. Phys. 47, 415 (1975).

[3] T. Alvesalo, T. Haavasoja, P. C. Main, M. T. Manninen, J. Ray, and L. M. M. Rehm, Phys. Rev. Lett. 43, 1509 (1979); T. A. Alvesalo, T. Haavasoja, M. T. Manninen, and A. T. Soinne, Phys. Rev. Lett. 44, 1076 (1980).

[4] P. R. Roach, M. W. Meisel, and Y. Eckstein, Phys. Rev. Lett. 48, 330 (1982).

[5] D. S. Greywall and P. A. Busch, Phys. Rev. Lett. 49, 146 (1982).

[6] S. Fantoni, V. R. Pandharipande, and K. E. Schmidt, Phys. Rev. Lett. 48, 878 (1982).

[7] G. E. Brown, C. J. Pethick, and A. Zaringhalam, to be published.

[8] G. E. Brown, V. Mishra, and C. J. Pethick, to be published.

[9] E. Krotscheck and R. A. Smith, to be published.

[10] C. H. Aldrich, III, and D. Pines, J. Low Temp. Phys. 32, 689 (1978).

[11] J. W. Clark and E. Feenberg, Phys. Rev. 113, 388 (1959).

[12] E. Feenberg, *Theory of Quantum Fluids* (Academic, New York, 1969).

[13] A. Fetter and J. D. Walecka, *Quantum Theory of Many-Body Systems* (McGraw-Hill, New York, 1971).

[14] J. P. Blaizot and B. L. Friman, Nucl. Phys. A372, 69 (1981).

[15] S. Engelsberg and P. M. Platzman, Phys. Rev. 148, 103 (1966).

[16] J. Lindhard, K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 28, No. 8 (1954).

[17] J. W. Clark, in *The Many-Body Problem*, edited by R. Guardiola and J. Ros, Lecture Notes in Physics Vol. 138 (Springer-Verlag, New York, 1981), p. 184; D. G. Sandler, N.-H. Kwong, J. W. Clark, and E. Krotscheck, in *Recent Progress in Many-Body Theory*, edited by J. Zabolitzky, Lecture Notes in Physics Vol. 142 (Springer-Verlag, New York, 1981), p. 228; J. M. C. Chen, J. W. Clark, and D. G. Sandler, Z. Phys. A 305, 223 (1982).

[18] E. Krotscheck, R. A. Smith, J. W. Clark, and R. M. Panoff, Phys. Rev. B 24, 6338 (1981).

[19] E. Krotscheck, Phys. Rev. A 26, 3536 (1982).

[20] R. A. Aziz, V. P. S. Nain, J. C. Carley, W. L. Taylor, and G. T. McConville, J. Chem. Phys. 70, 4330 (1979); R. Alrichs, P. Penco, and G. Scoles, J. Chem. Phys. 19, 119 (1976).

[21] J. P. Jeukenne, A. Lejeunne, and C. Mahaux, Phys. Rep. 25C, 83 (1976).

[22] W. G. Stirling, R. Sherm, P. A. Hilton, and R. A. Cowley, J. Phys. C 9, 1643 (1976).

[23] D. Pines, private communication.

[24] K. Skjold, C. A. Pelizzari, R. Kleb, and C. E. Ostrowski, Phys. Rev. Lett. 37, 842 (1976).

Two-Dimensional Plasma Resonances in Positive Ions under the Surface of Liquid Helium

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Plasma resonances have been observed in a two-dimensional sheet of positive ions held just under the surface of liquid ^{4}He. The ion effective mass deduced from the resonant frequencies is found to be weakly temperature dependent, increasing linearly with temperature. Strong nonlinear effects are observed in the wave propagation. The ponderomotive force of the plasma wave induces a static $2k$ distortion of the charge density, which produces large shifts of the resonant frequency with drive amplitude.

PACS numbers: 67.40.Yv, 73.90.+f, 52.35.Mw

The propagation of longitudinal plasma waves in two-dimensional single-component plasmas has been observed in several physical systems: electrons above the helium surface,[1] metal-oxide-semiconductor field-effect transistor inversion layers, and semiconductor layered structures.[2]

In this Letter we report the first observation of plasma resonances in yet another system: positive ions held just under the surface of liquid helium. The ions consist of singly ionized He atoms surrounded by a ~ 12-Å-diameter clump of solidified He atoms, which result from the electrostrictive pressure of the charge (the Atkins "snowball" model[3]). The combined action of the repulsive image potential and a vertical electric field of 100 V/cm is used to hold the ions at an equilibrium distance of 300 Å under the liquid-helium surface. The mean spacing between ions is several microns for the ion number densities used ($n_0 \sim 7\times10^7/\mathrm{cm}^2$), and hence they form a single two-dimensional layer at the surface.

The ions differ from the similar case of electrons above the surface in two major respects: The ions have a very large effective mass, $m^* \sim 40$ helium atom masses, and they are in strong thermal contact with the liquid. The higher mass leads to much lower resonant plasma frequencies, 100 kHz versus ~ 100 MHz for the electrons. The good thermal contact allows us to observe intrinsic nonlinear effects in the plasma-wave propagation. In contrast, the electrons above the surface are only weakly coupled to the liquid (through ripplon interactions) and nonlinear effects are primarily due to heating.[4]

Figure 1 shows a schematic cross section of the experimental cell, which is mounted on a dilution refrigerator. The geometry is cylindrical, with the ions held at the liquid surface between the lower electrode SG, which is positively biased, and the two upper electrodes A and C, which are grounded. The distance between the top and bottom electrodes is $h = 3.0$ mm. The circular electrode C, of radius 0.5 cm, is surrounded by the annular electrode A. A circular guard ring G of radius $R' = 1.5$ cm confines the charge at the edges of the cell. The liquid ^{4}He in the cell (which has been purified of residual ^{3}He by passing it through a Vycor superleak) is generally set with the liquid level a distance $d = 1.4$ mm above SG, as measured with a capacitance bridge. The liquid surface is charged[5] by briefly applying + 1600 V to a tungsten field emission tip just below a mesh grid in SG, and the charge can subsequently be held at the liquid surface for hours. The surface charge is measured at the end of the experiment by connecting an electrometer to the plates A and C and measuring the net induced charge that leaves the upper plates as the voltage on SG is turned to zero. The effective radius R of the charge pool is found following the analysis of Lambert and Richards,[6] and this allows the ion density n_0 to be calculated.

To generate the plasma waves a small ac voltage is applied to G, and the frequency is swept through the resonance. The signal is detected using a preamplifier to measure the voltage induced on C from oscillations in the surface charge density. The bottom curve in Fig. 2 is an experimental spectrum showing the first two resonant modes for an ion density $n_0 = (6.9 \pm 1)\times10^7/\mathrm{cm}^2$ at $T = 0.22$ K. A drive voltage of only 4 mV peak to peak on G is sufficient to give a large signal, with a signal-to-noise ratio of order 50. The signal can be observed at temperatures up to 0.47 K where the quality factor Q of the mode approaches one.

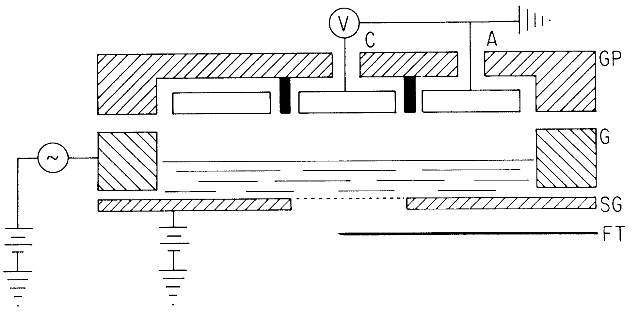

FIG. 1. Schematic cross section of the experimental cell, where the sheet of positive ions is held 300 Å below the helium surface.

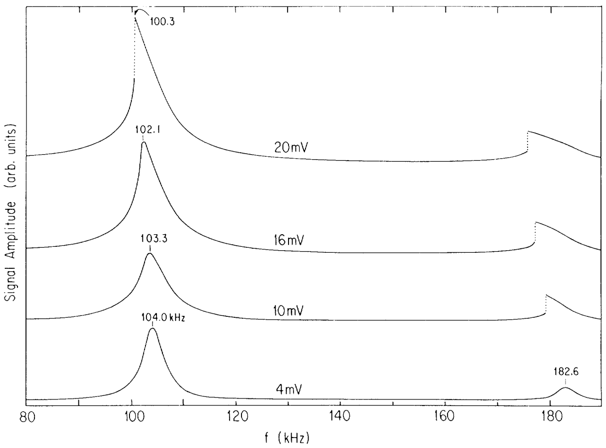

FIG. 2. Plasma resonance spectra at T = 0.22 K, displaying the resonance frequency shifts with increasing drive amplitude in millivolts peak to peak. $V_{SG} = +30$ V, $V_G = +35$ V.

For a comparison of these experimental resonant frequencies with theory, the equations describing the plasma motion, with neglect of dissipation, are[7,8]

$$\partial n/\partial t + \nabla \cdot (n\vec{v}) = 0 \,, \tag{1}$$

$$\partial \vec{v}/\partial t + (\vec{v}\cdot\nabla)\vec{v} = -(e/m^*)\nabla\varphi \,, \tag{2}$$

$$\nabla^2\varphi = -4\pi e n \delta(z-d) \,, \tag{3}$$

which are respectively the continuity equation, the acceleration equation, and Poisson's equation. $n(\vec{r},t)$ and $\vec{v}(\vec{r},t)$ are the local density and velocity of the ions, and $\varphi(\vec{r},t)$ is the electrical potential, which is subject to the boundary condition $\varphi = \text{const}$ at the bottom and top electrodes $z=0$ and $z=h$. For the cylindrical geometry the linearized radial solutions of (2) and (3) have the form $v(r,t) = \delta v J_1(kr)e^{-i\omega t}$, $n(r,t) = n_0 + \delta n J_0(kr)e^{-i\omega t}$, where J_1 and J_0 are Bessel functions. Inserting these in Eqs. (1)–(3) gives the linear dispersion relation

$$\omega^2 = (2\pi n_0 e^2 k/m^*)F(k) \,, \tag{4}$$

with[9]

$$F(k) = 2\sinh kd \, \sinh[k(h-d)]/\sinh kh \,. \tag{5}$$

The wave number k is determined[1,9] from a boundary condition $v=0$ at the charge radius $r=R$, and this gives $k=\alpha_n/R$ where α_n is the nth zero of J_1. For the first two modes in our cell $k_1 = 2.7$ cm^{-1}, $k_2 = 5.0$ cm^{-1}. We have observed the first five radial modes, and the frequency ratios agree with Eq. (4) to within few percent.

With use of the low-amplitude resonant frequencies from Fig. 2, Eq. (4) can be solved for the ion effective mass at 0.22 K. Both resonances yield $m^* = (37\pm 2)m_4$, which is somewhat smaller than the value $(43.6\pm 2)m_4$ found at 0.7 K by Poitrenaud and Williams.[10] In fact, we find that our resonant frequencies are temperature dependent, with a linear decrease of 5% between 0.1 and 0.4 K. If this is interpreted as a increase in the effective mass (since all the other terms in Eq. (4) should be temperature independent) then our results extrapolate fairly well to the Poitrenaud and Williams result at 0.7 K. Figure 3 shows the values of m^*/m_4 deduced from the frequency shift.

This temperature dependence of the effective mass, although weak, is still unexpected. In the Atkins model[3] of the positive ion the effective ion radius is determined by equating the electrostrictive pressure from the charge to the sum of the melting and surface tension pressures,

$$\frac{\alpha e^2}{2v_l r_+^4} = p_m + \frac{2\sigma_{ls}}{r_+}\frac{v_l}{v_l - v_s} \,, \tag{6}$$

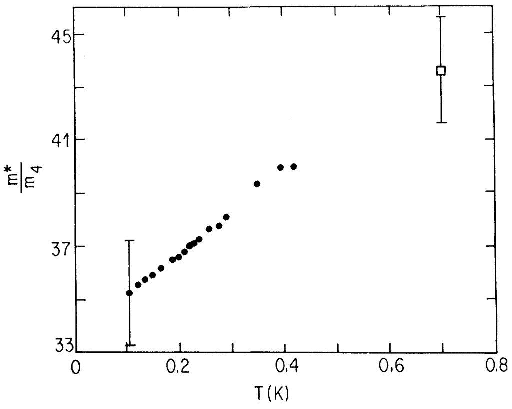

FIG. 3. Temperature dependence of the positive-ion effective mass deduced from the resonant frequencies. The open square is from Ref. 10.

where α is the molar polarizability, r_+ the ion radius, σ_{ls} the liquid-solid surface tension, and v_l and v_s the liquid and solid molar volumes [crudely approximated as being pressure independent in Eq. (6)]. The effective mass can be computed[10] from $m^* = [\frac{4}{3}\pi(4/v_s) + \frac{2}{3}\pi(4/v_l)]r_+^3$. Below 1 K the melting pressure p_m is constant, and only σ_{ls} could be temperature dependent in this model. However, the surface tension on bulk liquid-solid interfaces has recently been measured,[11] and no significant temperature dependence was observed (the expected $T^{7/3}$ term[12] is apparently very small). The 10% increase in effective mass we observe would require roughly a 12% decrease[13] in σ_{ls} as the temperature is raised from 0.1 to 0.4 K (corresponding to ~3% increase in r_+). It should be noted that the ion structure differs greatly from the bulk liquid-solid interface, with the "crystal surface" consisting of perhaps 20 atoms arranged on a sphere. The spectrum of surface excitations of the ion (which contributes a large portion of the surface energy) is substantially altered from the bulk modes. The density of states will be zero until wavelengths shorter than the ion circumference (~40 Å) are reached. As a result, σ_{ls} could be significantly different for the ion as compared with the bulk.

A significant aspect of our measurements is the strong nonlinear features that we observe. Figure 2 shows the result of increasing the drive amplitude. The resonances shift rapidly to lower frequencies, and can be fitted with a form $f^2 = f_0^2(1-\beta^2\varphi_G^2)$ where f_0^2 is the low-amplitude limit and φ_G is the drive voltage on G. For the first radial mode we find $\beta \cong 14$ V^{-1}, and the shifts are even larger for the second mode in Fig. 2. When the frequency shift exceeds the linewidth, the resonance curve becomes hysteretic in the classic manner of nonlinear oscillators, indicated by the dashed jumps in Fig. 2. These spectra were all taken sweeping up in frequency; when sweeping down through the first resonance (not shown in Fig. 2) with a 20-mV drive the jump occurred at 99.7 kHz compared with the 100.3 kHz observed sweeping up. Figure 4(a) shows both up and down sweeps for a lower charge density ($4\times10^7/\text{cm}^2$) where the effect is even more pronounced.

A theory of these nonlinear effects has been formulated by Ikezi.[14] He finds that the principal nonlinear mechanism is the $(\vec{v}\cdot\nabla)\vec{v}$ term[8] in Eq. (2), which is commonly known as the ponderomotive force in plasma physics. The time-averaged component of this force leads to a static deformation of the charge density at twice the wave number of the plasma wave. The greatest depletion of charge occurs where the ion velocity is highest, and this leads to a lowering of the resonant frequency. Ikezi finds for a one-dimensional standing wave that the shift is given by $f^2 = f_0^2(1-\beta'^2\varphi_m^2)$ where φ_m is the maximum potential in the wave, and for our case of small vertical fields $\beta' = k/\sqrt{2\pi n_0 e}$. Using $k = 2.7\ \text{cm}^{-1}$, $n_0 \simeq 7\times10^7/\text{cm}^2$, and approximating $\varphi_m \simeq Q\varphi_G$ gives $\beta \approx Q^2\beta' \approx 30\ \text{V}^{-1}$. The one-dimensional calculation cannot really be directly compared to our experimental Bessel function resonances, but it is clear there is at least order-of-magnitude agreement. The $1/n_0^2$ dependence of the frequency shift can also be qualitatively seen by comparing the 9-kHz shift in Fig. 4(a) with the 4.3-kHz shift in the top curve in Fig. 2. More detailed measurements and analysis are currently in progress.

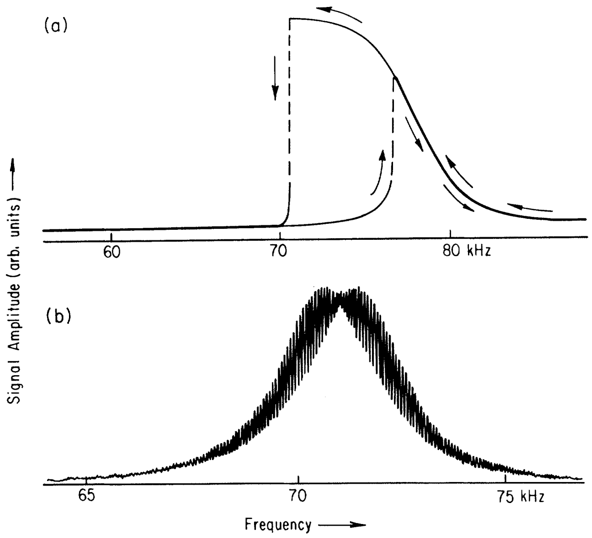

FIG. 4. (a) Example of the nonlinear hysteresis in the plasma resonance; $n_0 = 4\times10^7/\text{cm}^2$, $T = 0.18$ K, 20-mV drive. (b) Amplitude modulation of the plasma resonance due to oscillations of the helium surface.

When the level of mechanical vibrations in the experimental cell is increased (by readjusting vibration isolaters in the pumping line to the refrigerator's mechanical pump) a further effect of amplitude modulation can be observed in the plasma wave. Figure 4(b) is an example of this, showing a frequency sweep from 60 to 80 kHz which took 50 sec to complete. Readily visible is an amplitude modulation of the peak at a frequency of 5.6 Hz. This is the frequency of the first radial gravity wave of the cell, which was observed to be the dominant mode in a low-frequency fast Fourier transform spectrum of the charged-surface oscillations. Because of the vertical electric fields on the charge, the mounds and troughs of the gravity wave produce a distortion of the charge density at the wave number $k_g \approx k_1$, and on every half-cycle of the gravity wave the distortion reverses. This causes the plasma-wave resonant frequency to shift up and down about its equilibrium value, leading to the amplitude modulation at the gravity-wave frequency seen in Fig. 4(b).

In summary, we have observed the plasma mode in positive ions under the helium surface. The mode is able to probe details of the ion structure, and should be very useful in further measurements of the ion-surface and ion-ion interactions. The nonlinear effects should be of interest to plasma physicists, since although the plasma is extremely cold, the confinement time is infinite on their scale, and this allows a very detailed examination of the nonlinearities.

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[1] C. C. Grimes and G. Adams, Phys. Rev. Lett. 36, 145 (1976).

[2] For a comprehensive review, see T. Ando, A. B.

Fowler, and F. Stern, Rev. Mod. Phys. 54, 437 (1982).

[3] K. R. Atkins, Phys. Rev. 116, 1339 (1959); K. W. Schwarz, Adv. Chem. Phys. 33, 1 (1975).

[4] C. C. Grimes, Surf. Sci. 73, 379 (1978).

[5] A curious effect: Positive charge can accumulate at the liquid surface without the field tip ever being turned on (~10^5 charges in several hours)! We believe this is due to cosmic-ray ionization; see W. L. Willis, Los Alamos Report No. LA-DC-7026, 1965 (unpublished).

[6] D. K. Lambert and P. L. Richards, Phys. Rev. B 23, 3282 (1981).

[7] A. L. Fetter, Ann. Phys. (N.Y.) 81, 367 (1973).

[8] K. Mima, H. Ikezi, and A. Hasegawa, Phys. Rev. B 14, 3953 (1976).

[9] C. C. Grimes and G. Adams, Surf. Sci. 58, 292 (1976).

[10] J. Poitrenaud and F. I. B. Williams, Phys. Rev. Lett. 29, 1230 (1972), and 32, 1213 (1974).

[11] J. Landau, S. G. Lipson, L. M. Määttänen, L. S. Balfour, and D. O. Edwards, Phys. Rev. Lett. 45, 31 (1980).

[12] A. F. Andreev and A. Ya. Parshin, Zh. Eksp. Teor. Fiz. 75, 1511 (1978) [Sov. Phys. JETP 48, 763 (1978)].

[13] σ_{ls} must vary at least as $\sigma_{ls} = \sigma_0 - A T^{1+\epsilon}$ with $\epsilon > 0$. Our observation of roughly linear behavior implies that ϵ is small, $\epsilon \sim 0.1$. With such a small ϵ, m^* would not show deviations from linearity (approaching zero slope at $T = 0$) until well below 10 mK.

[14] H. Ikezi, to be published.

Geometry and Electronic Structure of Cl on the Cu{001} Surface

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The atomic geometry of Cu{001}$c(2\times2)$–Cl has been determined by surface extended x-ray-absorption fine structure to consist of a simple Cl overlayer in fourfold Cu hollows with a (2.37± 0.02)-Å bond length. With use of this geometry, self-consistent electronic structure calculations were performed and compared with angle-resolved photoemission data, giving excellent agreement for the position and dispersion of several Cl-induced surface states and resonances. These results have resolved previously reported discrepancies for the Ag{001}$c(2\times2)$–Cl system.

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The relationship between geometric and electronic structure at surfaces has been an important theme of modern surface physics. In a recent study, the electronic structure of a half-monolayer of Cl adsorbed on the Ag{001} surface with centered (2 × 2) periodicity was computed for two geometric models, a simple overlayer model (SOM) with Cl occupying fourfold hollow sites and a mixed-layer model (MLM) resembling an epitaxial AgCl{001} plane.[1,2] Comparison with ultraviolet photoemission spectra[3] indicated that the MLM gave the best agreement. In contrast, a study of low-energy electron-diffraction (LEED) intensity spectra showed better agreement with theoretical intensities based on an SOM rather than an MLM.[4] The present study was motivated by this apparent contradiction between two widely used techniques of surface science.

The Cu{001}$c(2\times2)$–Cl system was chosen as the closest analog to Ag{001}$c(2\times2)$–Cl which we could presently study with the desired methods. Its geometrical structure was first determined by surface extended x-ray-absorption fine structure (SEXAFS),[5] a technique capable of discriminating between SOM and MLM geometries and of measuring highly accurate bond lengths. The exact geometry was then used as input to a self-consistent electronic structure calculation, which was carried out with sufficient resolution to permit comparison with angle-resolved photoemission spectra. (This degree of detail had not been attempted in the Cl-on-Ag study.) Finally, angle-resolved spectra were measured for a surface prepared exactly as in the SEXAFS measurement. Excellent agreement was obtained between peaks in the calculated and measured spectra, thereby establishing the soundness and consistency of our procedures. The bonding in the Cu{001}$c(2\times2)$–Cl system ultimately indicated modifications of the SOM geometry used in the initial electronic structure calculations of the Ag{001}$c(2\times2)$–Cl system and these modifications have resolved the essential contradiction.[1-4]

In the SEXAFS experiment, performed at the Stanford Synchrotron Radiation Laboratory, a clean Cu{001} surface was exposed to 15 ±5 lang-