

EXTRAORDINARY BEHAVIOR OF  ${}^4\text{He}$  ON HYDROGEN AND  
DEUTERIUM SURFACES

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*Using third sound resonance below 1 K, for  ${}^4\text{He}$  on solid  $\text{H}_2$  and  $\text{D}_2$  surfaces, we have found new phenomena. On  $\text{H}_2$  we find surface superfluidity for  ${}^4\text{He}$  coverages less than one layer, with the zero temperature Kosterlitz-Thouless (KT) transition extrapolated to a coverage of  $0.35 \pm 0.05$  layers. On both  $\text{H}_2$  and  $\text{D}_2$  we find a new line of transitions below the KT transitions. On  $\text{H}_2$ , at zero temperature, the new line of transitions starts at zero temperature at a coverage of  $0.6 \pm 0.1$  layers of  ${}^4\text{He}$  and has a slope of 0.58 (K/layer) of the universal KT slope. We also find, on  $\text{H}_2$ , two modes of surface wave propagation between the new transition and the KT transition. Below the new transition we also find anomalous third sound absorption which increases exponentially with temperature by five orders of magnitude.*

Introduction

When helium is weakly bound to a surface at low temperatures new phenomena are found both theoretically and experimentally. A quantum wetting model has been presented by Cheng, Cole, Saam, and Treiner<sup>1</sup> (CCST) which points to a prewetting transition for adsorbed  ${}^4\text{He}$ . Several experiments have been reported during the past year which indicate non-wetting of  ${}^4\text{He}$  on cesium<sup>2</sup>. Using CCST terminology for the surface potential, Cs has the shallowest well depth ( $D$ ) while  $\text{H}_2$  has the weakest van der Waals attraction ( $C_3$ ). Early work using third sound in  ${}^4\text{He}$  on an  $\text{H}_2$  substrate was carried out by J.G.Brisson, et al<sup>3</sup> for coverages greater

than two layers, above 0.45K to find a Van der Waals coefficient of  $21\text{K}+17\text{K}-8.5\text{K}\cdot\text{layers}^3$ . Work in the same system, using a torsional oscillator, was also carried out by P.W.Adams, *et al*<sup>4</sup>. The early work was outside the region of the observations reported here. If the new transition we report below involves a change in the normal fraction, the torsional oscillator method should, in principle, see it. This method has yet to show such a transition. We report here our discoveries, to date, for the behavior of <sup>4</sup>He on the weak binding H<sub>2</sub> and D<sub>2</sub> surface. A more complete discussion of the results to be presented here will be given elsewhere<sup>5</sup>.

### Experiment

Measurements were carried out with two different experimental configurations. The first, with which most of the data shown below were taken, is fully described in Ref.6 and Ref. 7. The substrate gas is admitted at 77K into a 3 m<sup>2</sup> sintered copper film reservoir which in turn is connected to a third sound resonator by a 20 cm length of capillary with a 125 μm id. The apparatus is cooled slowly during about ten hours, starting at 30K, to 4.2K to allow time for ortho-para conversion and coverage equilibrium. At 4.2K, we pump on the reservoir-resonator system for two days to remove any <sup>4</sup>He. Because of the weak binding of <sup>4</sup>He atoms to the H<sub>2</sub> surface<sup>8</sup> of 17K the last layer of <sup>4</sup>He is removed rapidly with an estimated characteristic time of 10 minutes. Also, the fitting by CCST<sup>9</sup> of their calculations to our third sound data require no zero offset indicating that our measurements begin with a clean H<sub>2</sub> surface. When we add measured amounts of helium gas at 4.2K we see an immediate response in the third sound resonator pressure indicating that the connecting capillary is not plugged with H<sub>2</sub>. In more recent work we also cool slowly to 2K where we anneal the H<sub>2</sub> substrate for one day. The substrate thicknesses given are an average coverage. We have also grown our H<sub>2</sub> substrates on a surface preplated with argon, as suggested recently by Leiderer's studies<sup>10</sup>. The results reported below are generally unaffected by these various procedures as long as the average H<sub>2</sub> coverage is greater than two layers. However, it is possible to modify our results by a quench cool.

The second experimental configuration uses the same resonator but the connecting capillary is disconnected from the reservoir and sealed. This experimental geometry is similar to that described in Ref. 11. Both the substrate

gases and  $^4\text{He}$  must be added at room temperature by diffusion through the thin plastic walls of the resonator with a typical time constant of five hours. Here we carried out our measurements on an average coverage of 30 layers of  $\text{H}_2$  within the resonator and calibrated our  $^4\text{He}$  coverages using the low temperature third sound resonant frequency. With the obvious difficulty of this method there is a major advantage. We are using a small simply connected geometry which eliminates questions of different KT transitions in the film reservoir, U-tube oscillations between the resonator and reservoir, and  $\text{H}_2$  coverage in the resonator.

To test the reservoir-resonator apparatus for spurious third sound modes and transitions we have used third sound to investigate  $^4\text{He}$  on the well studied argon surface, with no intervening  $\text{H}_2$  substrate. The results are similar to earlier measurements, with no extra modes of sound propagation found, and only the KT transition seen. We find the KT transition line to have a slope within one percent of the KT slope for helium on  $\text{H}_2$  and we use this slope to establish a coverage scale. This is in contrast to our earlier work<sup>1,2</sup>, where we used a BET analysis of our adsorption measurements. As pointed out by Brisson<sup>3</sup>, the approximations of the BET method will be incorrect for adsorption in the quantum limit. Here we relate our KT slope to that found using torsional oscillators by the Cornell group<sup>1,3</sup>. The fifteen percent uncertainty in our coverage scale comes from the uncertainty in their superfluid coverage at the KT transition temperature ( $T_{\text{KT}}$ ) from which we have obtained a correction factor of  $0.75 = n_s(T_{\text{KT}})/n_s(0)$ , (see Eq.1). This uncertainty raised to the third power leads to an uncertainty in our Van der Waals coefficient for  $^4\text{He}$  on  $\text{H}_2$  of  $12\text{K}+6\text{K}-5\text{K}\cdot\text{layers}^3$ . A one parameter fit of our third sound measurements at low temperatures on  $\text{H}_2$  by CCST<sup>9</sup> to their model gives excellent agreement but produces a coverage scale 35% less than ours, beyond the range of our uncertainty. We find that a dose of  $40 \mu\text{moles}$  of helium corresponds to one layer, or  $13 \mu\text{moles}/\text{m}^2$ , in our helium film reservoir.

## Results

Fig.1 illustrates measurements of third sound resonance for various coverages. Velocity can be found by multiplying resonant frequency by 4 cm. We find two characteristic temperatures. The data ends as expected at  $T_{\text{KT}}$  where the resonance broadens and weakens with the loss of superflow. We also find a new characteristic temperature

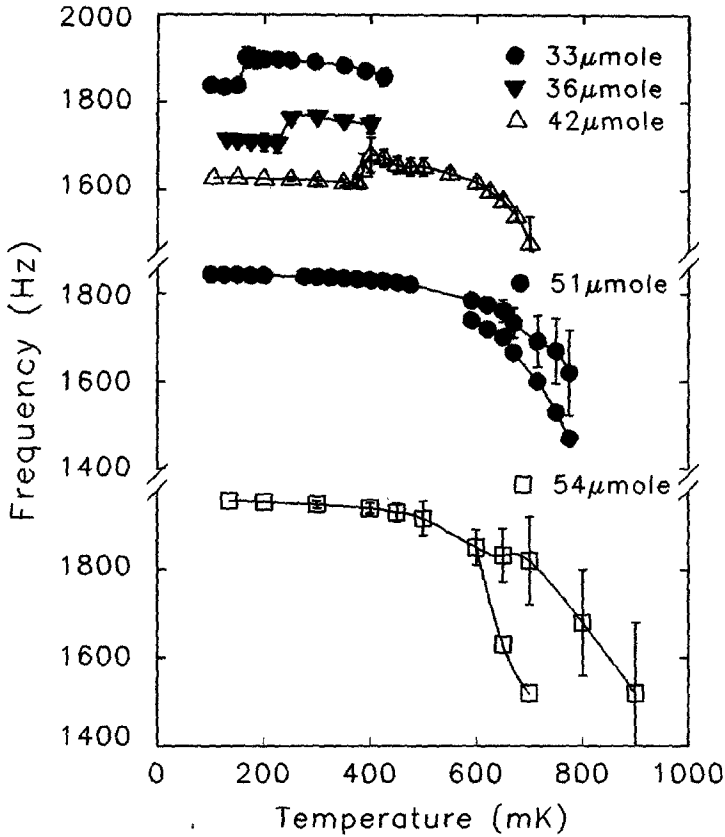


Fig. 1. Third sound resonant frequency for indicated dose of  ${}^4\text{He}$  on  $\text{H}_2$ . The  $51 \mu\text{mole}$  dose measurements were carried out in a sealed resonator with no reservoir.

( $T_c$ ) where there is a sharp jump in the resonant frequency for coverages at and below one layer. At larger coverage a new, second mode of surface wave propagation appears at  $T_c$ , with no sharp jump found in the third sound mode. Recently, we have carried out a detailed study of this new mode in our sealed resonator geometry and find that near the transition it has a phase differing by  $100 \pm 10$  degrees from the upper third sound mode, a line width 50 times larger, and an oscillator strength 50-90 times larger. With increasing temperature the two modes become more similar, their phase difference decreases, and they broaden to become indistinguishable at  $T_{KT}$ .

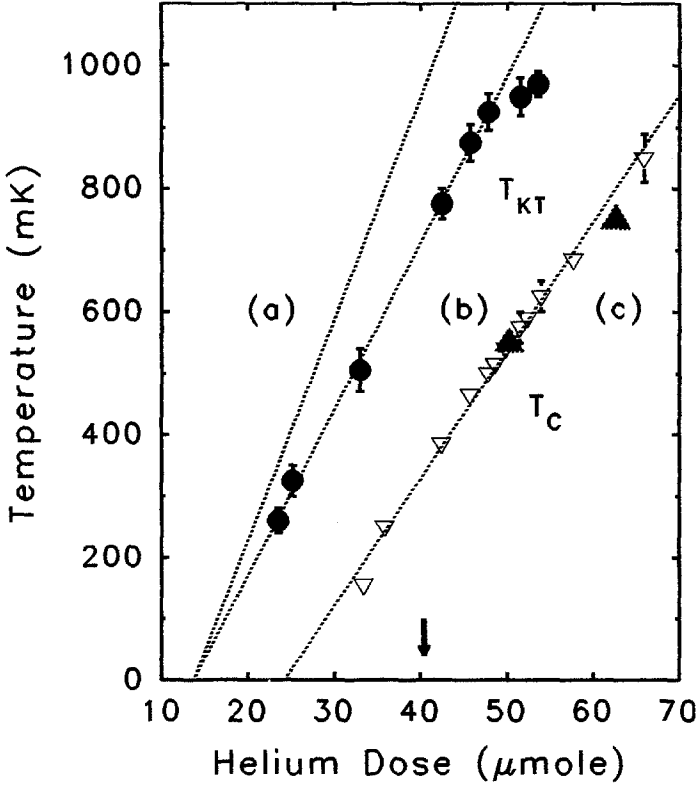


Fig. 2. Phase diagram from the two transitions shown in Fig. 1. The line of new transitions has a slope 0.58 of the corrected KT line. The two filled triangles are from the sealed resonator with no reservoir.

In Fig.2, we plot these two temperatures versus <sup>4</sup>He dose to obtain a phase diagram. The dotted line, without points, is related to the adjacent line of KT transitions with its coverages reduced by 0.75, a factor obtained from the Cornell data<sup>13</sup> as described above. This line should have the slope given by the universal KT relation<sup>14</sup>.

$$n_s \lambda_{KT}^2 = 4 \quad \text{where} \quad \lambda_{KT} = \left( \frac{h^2}{2\pi m k T_{KT}} \right)^{1/2} \quad (1)$$

where  $\lambda_{KT}$  is the deBroglie thermal wavelength evaluated at  $T_{KT}$  and  $n_s$  is the number of superfluid atoms per unit area

just before the transition. The intercept at zero temperature indicates the onset of superfluidity at a dose of  $14 \mu\text{moles}$  or 0.35 layers on  $\text{H}_2$ . The line of new transitions has a slope of 0.58 of the KT slope (the constant 4 in Eq.1 would be replaced by 6.9) and a zero temperature intercept at a dose of  $24 \mu\text{moles}$  or 0.6 layers. All coverages have an uncertainty of 15% from the factor of 0.75 as discussed above. The presence of two modes in region (b) of Fig. 2 indicates an additional degree of freedom. Further discussion can be found in Refs. 5.

It is natural to extend these measurements to a  $\text{D}_2$  substrate. Using similar methods we obtained the preliminary data shown in Fig. 3. In the limit of low coverage of  $^4\text{He}$  the third sound velocity is within 1% of that found on  $\text{H}_2$ , at multilayer coverage the velocity is 10% larger than on  $\text{H}_2$ . Most of this increase is expected due to

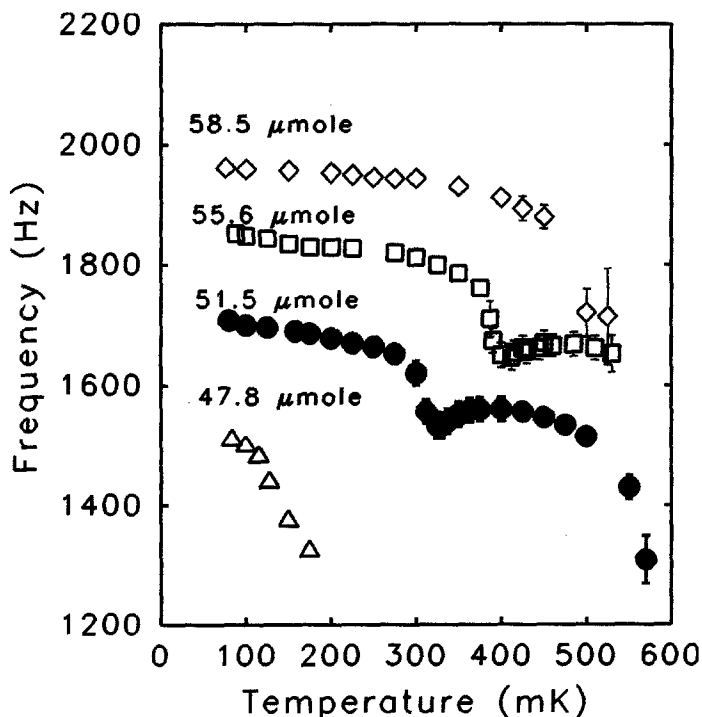


Fig. 3. Third sound resonant frequencies versus temperature at constant indicated dose of  $^4\text{He}$  on a  $\text{D}_2$  substrate. The anomalies are at higher coverage than on a  $\text{H}_2$  substrate.

the higher molecular density of  $\text{D}_2^{15}$ . As seen in Fig. 3 there is again a new set of transitions. The shape is different and we have yet to find a second mode. In Fig. 4 we generate the phase diagram for the new transitions and KT transitions on  $\text{D}_2$ . The slopes are similar to the  $\text{H}_2$  data - although more data must be taken for an accurate comparison. There is also a general displacement of 12-14  $\mu\text{moles}$  to higher  ${}^4\text{He}$  coverage for the zero temperature intercepts giving 0.6 layers for the KT transition and 0.95 layers for the new transition.

During these investigations we noticed large variations in third sound absorption. To investigate absorption as a function of temperature we increased our  ${}^4\text{He}$

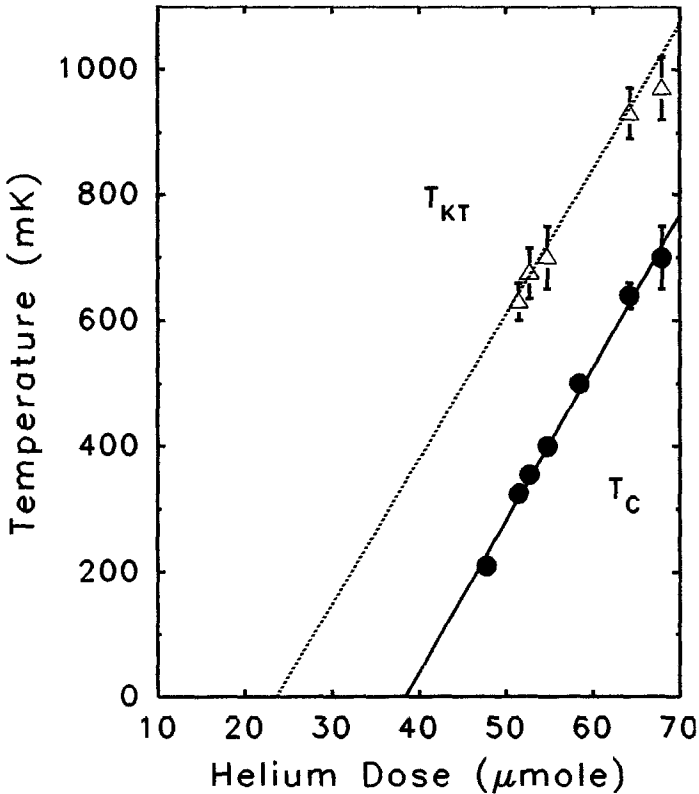


Fig. 4 The phase diagram of the transitions in  ${}^4\text{He}$  on  $\text{D}_2$  shown in Fig. 3. Compared to Fig. 2 the transitions are shifted upward in coverage by 12  $\mu\text{moles}$  (or 0.3 layers).

dose on  $H_2$  to 91  $\mu$ moles thus moving both transitions above 1.2K, outside the range of measurement. Shown in Fig.5 is the inverse Q, which when multiplied by the resonator length (4 cm) gives the absorption length, plotted against temperature. There is an exponential rise in absorption through five decades with a characteristic temperature,  $T_0$ , of 65mK. If the rise had continued through the last decade we would have lost superflow at 0.9K (for 1200Hz), a factor of two below  $T_{KT}$ . Typical behavior on stronger binding substrates, such as argon, generally produce an inverse Q near  $10^{-4}$  with no more than a factor of ten increase until the KT transition is approached. On the  $H_2$  surface, at our lowest temperatures of 70mK we find the smallest absorptions that we have seen on any substrate,  $1/Q$  less than  $10^{-5}$ . We

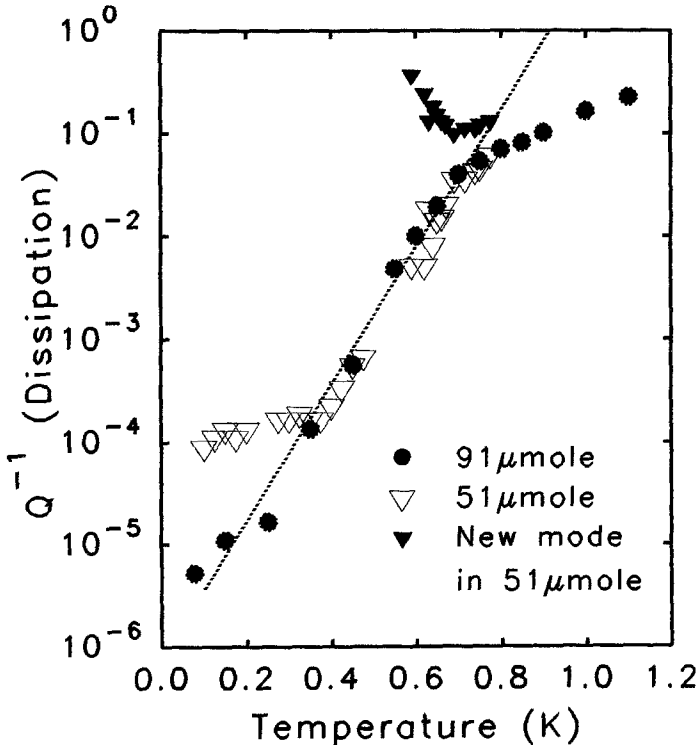


Fig. 5 Third sound absorption for two coverages. The filled triangles are the new mode which appears at 0.56K. The unusual exponential rise in absorption with temperature has a characteristic temperature of 65 mK.



are also carrying out measurements of absorption as a function of coverage. Eq.2 summarizes our initial absorption results for doses greater than 90  $\mu$ moles.

$$\frac{1}{Q} \propto \frac{e^{T/T_0}}{\sigma^{2-2.5}} \quad (2)$$

The dash indicates the range of the exponent of the coverage  $\sigma$ . We have also added to Fig.5 results from a low coverage measurement in our sealed resonator. In the limit of low coverage the third sound absorption becomes independent of coverage and the trend with temperature is not altered by the new transition. For the low coverage we have also added the absorption of the new mode of surface sound in Fig. 5.

The exponential increase with temperature is not generally found in nature, but has been seen in two recent experiments measuring the resistance in atomically thin superconducting films. Y. Liu, *et al*<sup>16</sup> at Michigan reported this behavior in lead films deposited at low temperatures appearing well below the bulk transition temperature. R.C.Dynes has reported similar results<sup>17</sup> for lead films only when deposited in a granular state.

### Conclusion

For this conference proceeding we have restricted the discussion and conjecture and simply reported the results. A new phase transition has been found starting in submonolayer coverages of <sup>4</sup>He on H<sub>2</sub> and D<sub>2</sub>. It is not clear at present whether it involves morphology and wetting, a new order parameter, or a surface interaction due to the large zero point motion of the substrate.

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