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Positive-Ion Structure in a Tricritical ^{3}He-^{4}He Mixture

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(Received 8 June 1978)

The mobilities of positive and negative ions have been measured in the normal phase of a ^{3}He-^{4}He mixture near the tricritical point. Whereas for negative ions nearly the same weak temperature dependence in observed as in pure ^{3}He, the mobility of positive ions in the mixture drops sharply by almost a factor of 2 when phase separation is approached. This is attributed to condensation of ^{4}He onto the positive-ion surface.

Ions have served as a very useful probe to study the properties of liquid helium, like recently the superfluid phases of ^{3}He.[1] The structure of these ions, both negative ("electron bubbles") and positive ("snowballs"), seems to be well understood.[2] Yet for a particular case, namely the mobility of positive ions in ^{3}He, experimental data have shown a surprisingly large scatter at temperatures around 0.2 K.[3-5] Roach, Ketterson, and Roach[6] and Bowley[7] ascribed this lack of reproducibility to minute traces of ^{4}He atoms, which condense on the snowball at low temperatures, leading to an increase in the radius of the positive ion and hence to a lower ion mobility. This modified structure of the positive ions in very dilute mixtures of ^{4}He in ^{3}He was investigated in more detail by Sluckin.[8] According to his theory phase separation occurs in the mixture in the vicinity of the ion, with a halo of ^{4}He-rich phase around the ion, separated from the surrounding ^{3}He-rich phase by an additional phase boundary. The halo radius R_H is predicted to grow as the temperature is lowered, until it reaches its final value at a temperature where macroscopic phase separation sets in the bulk liquid.

In a tricritical ^{3}He-^{4}He mixture, where the concentration of ^{4}He is considerably higher ($x_4 = 0.325$), one would expect a similar behavior. Since phase separation in such a mixture occurs at a much higher temperature ($T_t = 0.867$ K) than for the dilute mixtures studied so far, the halo too should form at higher temperatures, thus lowering the mobility of the positive ion in a temperature range where until now no anomalies have been observed.

We have investigated the ion mobility μ in such a tricritical mixture between 0.5 and 1.1 K. A time-of-flight method was used similar to that described by Ahonen *et al.*[1] The ion current from an ^{241}Am source was shaped into pulses by means of a gate grid, and the velocity of these ion packets in a drift space was determined from the arrival time at the collector. The ion pulses were sampled by means of a fast electrometer and a signal averager. Since the drift space was located in the upper half of the sample cell, all data points were taken in the *normal* phase of the mixture; the ^{4}He concentration x_4 was constant above the tricritical point, and decreased along the ^{3}He-rich side of the phase diagram below T_t to $x_4 = 0.09$ at 0.5 K.

The results for the mobilities in this tricritical mixture are plotted in Figs. 1 and 2. For positive ions (Fig. 1), a prominent drop of μ_+ by 40% is observed as the tricritical point is approached from above. Below T_t the mobility shows but a small further decrease down to 0.5 K. In contrast, for negative ions no such step near T_t was found, as the data in Fig. 2 indicate. The measured mobilities were independent of the applied electric drift field E_D in the studied range, $5 \leq E_D \leq 80$ V/cm, and there was no indication for several species of positive charge carriers with different mobilities.[7,9]

A comparison of our data with earlier results for pure ^{3}He, and also for dilute mixtures of ^{4}He in ^{3}He (which do not deviate from pure ^{3}He for $T > 0.5$ K) lead to the following conclusions: For *negative* ions the mobility in the tricritical mixture—like in pure ^{3}He[2,10]—can be well described in terms of Stokes's law for temperatures above

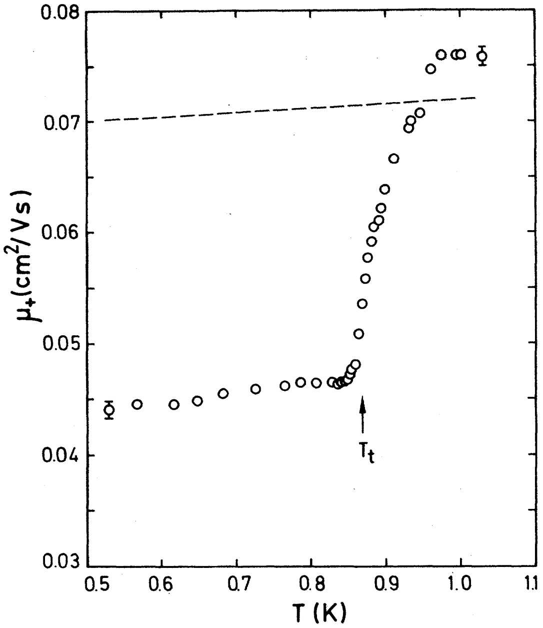

FIG. 1. Mobility μ_+ of positive ions vs temperature in a tricritical ^{3}He-^{4}He mixture at saturated vapor pressure (svp). For comparison, the mobility of positive ions in pure ^{3}He (Ref. 6) is represented by the dashed line. T_t denotes the tricritical temperature at svp. The uncertainty in the absolute value of the mobility is 10%.

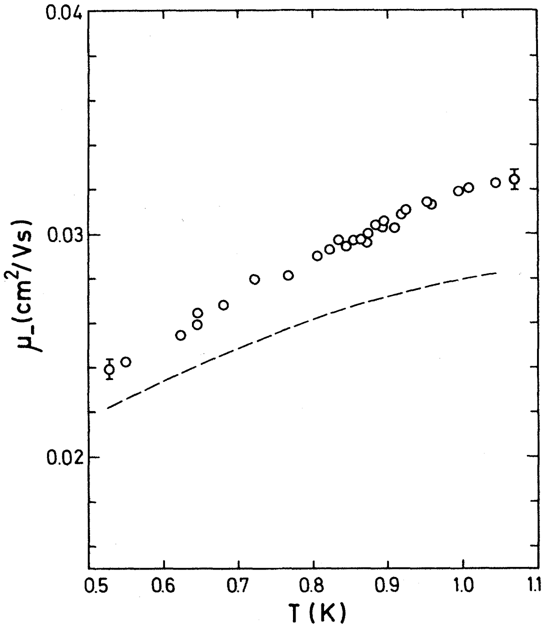

FIG. 2. Mobility μ_- of negative ions vs temperature in a tricritical ^{3}He-^{4}He mixture. The dashed line represents data of μ_- in pure ^{3}He (Ref. 3).

1 K:

$$\mu_- = e/4\pi\eta R_-, \qquad (1)$$

assuming for the viscosity $\eta \sim 20$ μpoise at 1 K[11] and for the radius of the negative ion $R_- = 19$ Å. Below 1 K μ_- varies less than the viscosity, because the mean free path of the quasiparticle excitations becomes comparable to the ion radius.[2] Since the ^{4}He concentration decreases below the tricritical point, the data for the mixture begin to approach the curve for pure ^{3}He, leading to a somewhat stronger temperature dependence of μ_- than in the case of pure ^{3}He. The large concentration fluctuations in the vicinity of the tricritical point[12] apparently have no measurable influence on the mobility.

For *positive* ions because of their smaller radii the inverval between 0.5 and 1.1 K is well below the validity of Stokes's law. In this temperature range quasiparticle scattering yields a more adequate description for the nearly constant mobility in pure ^{3}He.[2] If we now search for an explanation for the rapid decrease of μ_+ in the tricritical mixture, we might ascribe it either to a change in the structure and hence the size of the ion, or to a change of the properties of the surrounding liquid. The latter possibility can be ruled out, however, because the mobility μ_- of the negative ions should similarly be affected in this case, in contrast to the observed smooth behavior.

We therefore conclude that the mobility drop is due to a growth of the positive ion, probably connected to the phase separation occurring at the tricritical temperature. In comparison to T_t at saturated vapor pressure the step appears to be slightly shifted to lower temperatures, which we attribute to the elevated pressure near the ion surface.[13,2] The increase of the ion radius R_+ can be roughly estimated from the mobility, which in the region of quasiparticle scattering varies as $\mu \propto R^{-2}$ (neglecting the varying ^{3}He concentration along the coexistence curve).[2] This leads to a change of about 20% in R_+. Assuming that the radius far above T_t is $R_+ \sim 7$ Å, as in pure ^{3}He, we obtain $R_+ \sim 8.5$ Å below T_t. On the other hand the halo radius predicted for positive ions in the ^{3}He-rich phase of a phase separated mixture is approximately given by[8]

$$R_H \approx [\gamma(n_4 - n_3)/2\sigma_i]^{1/3}. \qquad (2)$$

Here n_4 and n_3 are the number densities of the ^{4}He- and ^{3}He-rich phase, respectively, σ_i is the interfacial tension of the mixture, and $\gamma = 1.91 \times 10^{-44}$ cgs units. For low $T < 0.3$ K, σ_i is nearly constant, and the predicted halo radius is $R_H = 13$ Å. At higher temperatures, where the interfacial tension decreases, a larger halo radius is expected from Eq. (2), e.g., $R_H = 15$ Å at 0.5 K and 30 Å at 0.8 K. In particular, on approaching the tricritical point from below, σ_i vanishes as $\sigma_i \propto t^2$,[14] and $n_4 - n_3 \propto t$,[15] where $t = 1 - T/T_t$. Hence according to (2) the halo radius ought to diverge as

$$R_H \propto t^{-1/3}. \tag{3}$$

Our data, however, do not show such a divergence below the tricritical point, but rather yield a temperature-independent value of the ion radius, which is larger by about 1.5 Å—i.e., less than 1 atomic layer—than R_+ at 1 K. One might argue that as the interfacial tension decreases and the postulated halo grows, the assumed spherical halo boundary might well be deformed, if not destroyed, by the ion moving at drift velocities of about 1 cm/sec. In this case, however, one would expect a field-dependent mobility, which was not observed. Besides, the drift velocities of the ions were much smaller than their thermal velocity. Therefore, provided the halo is not already destroyed by thermal motion, the drift of the ion should not destory it either.

Thus far the finite thickness L of the interface between the halo and the surrounding liquid has not been taken into account. The above treatment certainly has to be modified when L becomes comparable to the predicted halo radius. An estimate for the interfacial thickness can be obtained from the correlation length ξ for concentration fluctuations in the bulk, using the fact that $L \sim \xi$.[16] The correlation length in the ^{4}He-rich phase near the tricritical point was determined as[12] $\xi = 1.3xt^{-1}$ Å. A comparison with Eq. (2) shows that the condition $L \ll R_H$ is certainly not fulfilled in the asympotic tricritical region ($t < 0.01$). For temperatures further below T_t data for ξ are not available, but presumably in the whole range studied here the finite correlation length cannot be neglected in calculations of the positive-ion structure and mobility.

In summary, our experiment presents clear evidence for the condensation of ^{4}He onto the surface of positive ions, when phase separation is approached, but no indication for a halo of more than 1 atomic layer.

We are indebted to W. Schoepe for the loan of a fast electrometer and for many helpful suggestions, to H. Kinder and H. Meyer for valuable discussions, and to the Deutsche Forschungsgemeinschaft for financial support.

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