Microscopic Gas-Liquid-Like Phase Transition around the Positron in Helium Gases

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The clustering of atoms around slow positrons in helium gases is experimentally studied and the results are interpreted in terms of liquid drop formation. The critical temperature and pressure of the droplet phase are found to be 6.6 K, 5.4 atm in ^{3}He and 8.4 K, 7.6 atm in ^{4}He. The critical densities coincide with those of the helium liquids.

Charged particles exhibit abnormal behavior in low-temperature helium.[1] The repulsive exchange interaction between an excess electron and He atoms induces a cavity around a free electron as well as around an electron bound in a positronium atom.[2-5] On the other hand, attractive electrostatic forces cause clustering of He atoms around positive ions, which is assumed to lead to solid core ("snowball") formation in the liquid.[6] Also in the case of the positron, clustering effect have been reported in the gas phase,[5,7,8] whereas in the liquid phase mainly normal behavior is seen.[3,9] In this Letter we report experimental results showing that the electrostatic forces cause a gas-liquid-like phase transition around the positron well above the critical temperatures of the ordinary helium liquids.

When positrons from radioactive isotopes enter helium, they slow down by inelastic collisions and finally reach an equilibrium annihilation rate. By measuring this rate we directly obtain the density of helium in the immediate vicinity of the positron. The clustering of atoms is seen as a sudden increase of this equilibrium annihilation rate[5] in the positron lifetime spectrum. In this work the clustering is studied by measuring the annihilation rate of slow positrons in low-temperature helium gases as a function of pressure and temperature. From the results we extract various thermodynamic properties of these clusters using the analogy to the macroscopic system of condensing gas.

The helium chamber used in the experiment was a spherical 2-cm-diam cavity, where the temperature and pressure could be regulated with an accuracy of 0.01 K and 0.05 atm, respectively. The positron source, placed close to the wall of the chamber, was 3 μCi of NaCl on a thick gold foil. The conventional start-stop system for the lifetime measurement had a time resolution of 1.2 nsec.[3] The statistical error of the analyzed annihilation rates is less than 2% on the average. The necessary pVT data for ^{4}He are readily available.[10] Since no such data exist for ^{3}He we produced our own, accurate to about 3%.

Figure 1 shows the annihilation rate λ of slow positrons in ^{3}He and ^{4}He at different temperatures as a function of atomic density n in the chamber. At temperatures greater than 10 K we always see linear (or almost linear[9]) dependence of λ on helium density. However, at lower temperatures large deviation from this relationship is seen at densities $n \approx (0.1-2) \times 10^{22}$ cm^{-3}. At a certain relatively low density a distinct jump in the annihilation rate occurs indicating a sudden increase in the local helium density around the positron, i.e., the formation of a cluster. Once the cluster is formed, the annihilation rate, and thus the density of the cluster, stays practically constant (plateau region) independent of the bulk density of the gas, until it resumes the linear dependence. At this point the bulk density reaches that of the cluster and thus at higher densities the cluster effects are no longer visible. It is worth pointing out that the clustering occurs also in the liquid phase when its density is low enough.[9] With increasing temperature the cluster formation is

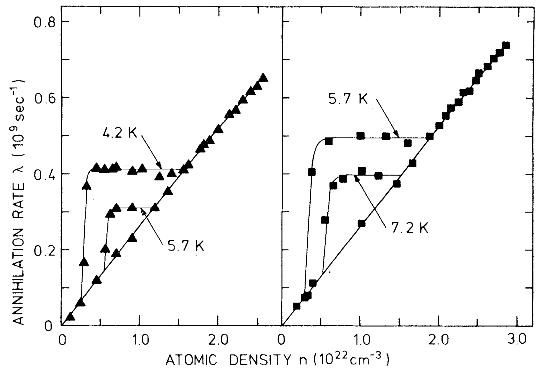

FIG. 1. The annihilation rate λ of slow positrons in ^{3}He (left) and ^{4}He (right) as a function of atomic density at different temperatures. The droplet formation is seen as a sudden jump from the linear dependence between λ and n.

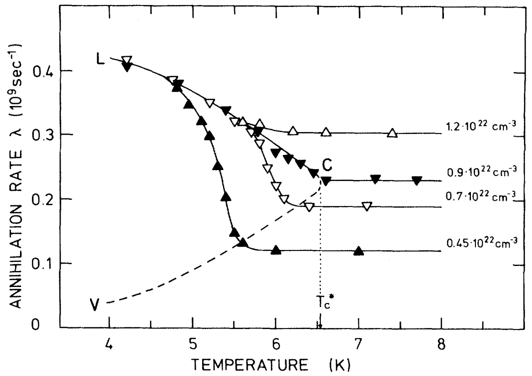

FIG. 2. The annihilation rate of slow positrons in ^{3}He gas as a function of temperature with the atomic density of the gas as a parameter. The critical point of the droplet phase is denoted by C. The curve LCV represents the liquid-vapor equilibrium curve of this phase.

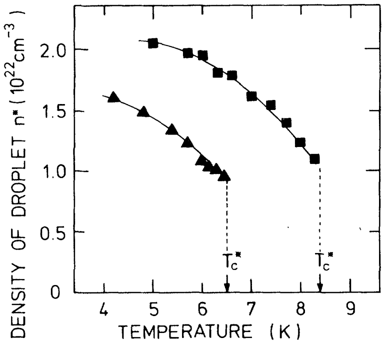

FIG. 3. The density of the droplet around the positron in ^{3}He (triangles) and ^{4}He (squares) gas as a function of temperature. Note the elevated critical temperatures (but practically the same critical densities) compared to those of the ordinary helium liquids.

shifted towards higher densities and the annihilation rate in the cluster decreases. Above a certain critical temperature, different for the two gases, all cluster effects disappear.

In Fig. 2 we show the annihilation rate λ as a function of temperature with ^{3}He density as a parameter. The annihilation rate stays constant above 6.6 K at fixed gas density (corresponding to the linear dependence in Fig. 1). At lower temperatures and low gas densities the transition into the cluster state is clearly seen. With increasing gas density the transition temperature increases and the change of λ in the transition decreases and finally disappears at $n = 0.9 \times 10^{22}$ cm^{-3}. Thus the point C represents a critical point beyond which no clustering occurs. In fact the behavior illustrated in Fig. 2 is analogous to that of condensing gas in a closed volume, the vertical axis corresponding to density. Accordingly we interpret the point C as the critical point of the cluster phase around the positron. The curve LCV corresponds to the liquid-vapor equilibrium curve and the parts LC and VC correspond to the liquid and vapor saturation curves, respectively. Thus we interpret the cluster around the positron as a liquid droplet. The critical temperature T_c^* of the droplet phase in ^{3}He is found to be 6.6 ± 0.1 K. The corresponding temperature for ^{4}He is 8.4 ± 0.1 K. This droplet formation occurs well above the critical temperatures T_c of the ordinary helium liquids, the ratio T_c^*/T_c being 2.0 and 1.6, respectively.[11] The form of the "liquid-vapor equilibrium curve" is also different; its shape near T_c^* is more pointed than for ordinary helium liquids. In addition, the transition is not discontinuous but has a finite width of the order of a few tenths kelvin. This feature, visible also in earlier measurements,[5] is evidently due to the finite number of particles in the droplet.

The height of the plateau in Fig. 1 directly gives the density n^* of the droplet. The results are shown in Fig. 3 as a function of temperature. The droplet densities are of the same order as densities of normal helium liquids at low pressures. The critical densities n_c^*, $(0.90 \pm 0.05) \times 10^{22}$ cm^{-3} and $(1.00 \pm 0.05) \times 10^{22}$ cm^{-3} for ^{3}He and ^{4}He respectively, coincide within experimental errors with those of the ordinary liquids (0.90×10^{22} cm^{-3} and 1.02×10^{22} cm^{-3}) giving further support to the droplet interpretation. Thus the effect of the electrostatic forces around the positron is mainly to cause local condensation above the bulk transition temperature, while the critical density of the condensed phase is not changed.

In this connection it should be pointed out that the useful analogy to a macroscopic system must not be taken too literally, i.e., the droplet density is evidently not quite uniform and so the annihilation rate actually gives an effective density, which is obtained by weighting the density profile of the droplet with the square of the positron wave function. However, the plateau region in Fig. 1 shows that this effective density stays constant although the bulk density and pressure change over a large interval.

The pressure needed for the droplet formation is obtained from the density values corresponding to the distinct jumps in Fig. 1. This pressure, shown in Fig. 4 as a function of tempera-

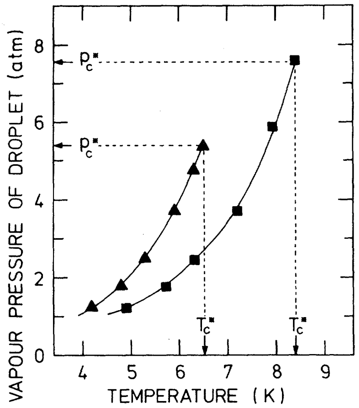

FIG. 4. The saturated vapor pressure (formation pressure) of the droplet around the positron as a function of temperature in ^{3}He (triangles) and ^{4}He (squares) gas. The high values of the critical pressures are due to the elevated critical temperatures.

ture, is analogous to the saturated vapor pressure of condensing gas. As is expected, in the overlapping temperature region the vapor pressure curves of the normal liquids seem to lie above those of the droplets, i.e., the condensation around the positron occurs at lower pressures than the bulk condensation. The relatively high critical pressure values for the droplet (5.4 ± 0.2 atm and 7.6 ± 0.2 atm for ^{3}He and ^{4}He, respectively) are simply due to the elevated critical temperatures.

Since the positron is a light and relatively weakly localized particle the electrostatic forces around it cause droplet formation in the gas phase but no solid core is formed in the liquid phase.[3] This fact is also supported by our preliminary measurements in liquid and solid helium, which show that the discontinuity of the annihilation rate due to solid formation around the positron occurs at the solidification pressure of the bulk liquid.

Presumably the size of the droplet is of the same order (radius 10–20 Å) as density inhomogeneities induced by other charged particles and it evidently depends on the temperature and pressure. The fluctuations due to the finite size, appearing as smearing of the transitions in Figs. 1 and 2, increase considerably near the critical points. Surprisingly, the fluctuations seem to be clearly larger in ^{4}He than in ^{3}He. These features remain to be investigated later.

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