Magnetic Susceptibility of ^{3}He Adsorbed on Sintered Silver Powder

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The magnetic susceptibility of ^{3}He adsorbed on sintered silver powder has been measured in the temperature range from 0.8 mK to about 10 mK with a SQUID magnetometer for eight different film thicknesses between monolayer and 15 layers. The measured susceptibility of films less than about 5 layers thick obeys the Curie law with the Curie constants given by the free localized model. In films more than 5 layers thick, the measured susceptibility is described by the Curie-Weiss behavior with a Curie-Weiss temperature of about 0.4 mK.

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Over the past several years, it has been revealed that ^{3}He layers adsorbed on a solid substrate at low temperatures play an important role in phenomena such as the surface-induced ferromagnetism of liquid ^{3}He,[1-4] the strong magnetic coupling between substrate nuclear spins and adsorbed ^{3}He spins,[5-7] and the thermal boundary resistance between sintered silver powder and liquid ^{3}He.[8] The underlying mechanism of all these phenomena has not yet been clarified, though several theoretical interpretations of the surface-induced ferromagnetism have been published.[9-10] It is clear that ^{3}He film adsorbed on a solid substrate provides a new system which can exhibit quantum mechanical phenomena. However, no experimental work has yet been published on ^{3}He adsorbed on a solid substrate at ultralow temperatures without bulk liquid ^{3}He present. This report presents the first measurement of the static magnetization of ^{3}He adsorbed on the surface of sintered silver powder as a function of the film thickness in the temperature range between 0.8 and 10 mK. In this experiment, we could succeed in measuring at what film thickness the ferromagnetic interaction becomes important, how many layers contribute to the ferromagnetism, and how the Fermi-degenerate liquid on the film affects the interaction.

The static magnetization of ^{3}He adsorbed on sintered silver powder was measured by a superconducting quantum interference device (SQUID) magnetometer.[11] A field of 480 Oe was applied throughout all the measurements presented here. The sample cell was machined from a rod of 99.99% pure silver, and the substrate consisted of 99.999% pure pulverized silver shot.[12] The purity of the adsorbed ^{3}He was specified by the manufacturer to be 99.999%.

The fine silver powder was "presintered" at 200 °C for 20 min in 3 Torr of H_2 gas, and was then ground with a mortar and pestle. A scanning electron microscope photograph of the resulting powder indicated the average particle size to be about 1000 Å. The presintered powder was then compressed tightly in a layer 1 mm thick at the bottom of the sample cell. The additional (nonpresintered) powder was packed on top of the 1-mm-thick layer of presintered powder by a hand press to a packing factor of 44% and was kept at 130 °C for 5 min, during which the powder was kept under pressure by a Teflon jig.[13] The mass of silver powder was 5.09 g per side. The surface area of the silver powder after the final sintering was measured by the Brunauer-Emmett-Teller method using N_2 gas at 77 K, and was found to be 5.15 m^2 per side. The surface area per unit mass obtained here is close to another recently reported value.[14]

The number of layers of the adsorbed ^{3}He film was then calculated from the measured surface area and the number density n of the films. For monolayer coverage, we adopted the value $n = 0.11$ Å^{-2} from the data of McCormick, Goodstein, and Dash[15]; for the second layer, we used the value given by Brewer[16]; and for thicker films, we used the density of saturated-vapor-pressure liquid.[17]

The sample cell was cooled by a single-stage copper nuclear adiabatic demagnetization apparatus. The temperature was measured by the free induction decay of platinum NMR[18] and was calibrated using a U. S. National Bureau of Standards superconductive fixed-point device (SRM-768).

The experiment was carried out as follows. The sample cell was thoroughly flushed with dry nitrogen at room temperature before being cooled. The background magnetization signal of the empty sample cell was measured down to 0.8 mK and was found to be weakly temperature dependent. The measured background magnetization was reproducible within the precision of our measurement even after the temperature of the sample cell had been raised to 10 K. A monolayer equivalent of ^{3}He gas was then introduced into the cell which was maintained at around 5 K for more than 8 h for "annealing" before cooling to lower temperatures. The cell was then cooled to 1 mK, and the total magnetization signal was measured as a function of temperature. The temperature dependence of the contribution of ^{3}He to the magnetization for a given thickness was computed by subtracting the background magnetization from the total magnetization. When ad-

ditional ^{3}He was introduced, the sample was again annealed in the same way as before. This procedure was repeated up to 15 layers of ^{3}He. Finally the cell was filled with saturated-vapor-pressure ^{3}He, and its magnetization was measured.

Because of the dc measurement technique used with SQUID, there is an ambiguity in the zero magnetization reference for both the background and the total magnetization signals. We therefore analyzed our data by fitting them with the Curie-Weiss law using the following equation:

$$V_T - V_B = (AH)/(T-\theta) + B \equiv M + B, \quad (1)$$

where V_T, V_B, H, and T denote the total and the background magnetization signals, the externally applied magnetic field, and the temperature, respectively; and A, θ, and B are fitting parameters. Parameter A is proportional to the Curie constant for a given film. The low-temperature magnetization of degenerate liquid ^{3}He is contained in parameter B in Eq. (1).

The conversion factor for converting from measured signal voltage to magnetization, calculated with the transformer characteristics of our SQUID magnetometer, was 1.06×10^{-8} emu/mV.

The open circles in Fig. 1 show the inverse of the temperature-dependent part of the measured magnetization of 1, 2, 5, and 8.5 layers of adsorbed ^{3}He. Not all the film thicknesses measured are shown in this figure. The straight lines represent the fit of Eq. (1) to the temperature-dependent part of the data. The slopes of the lines give the values of AH, and the intercepts give the values of θ. As can be seen from the figure, the magnetization obeys the Curie law up to a film thickness of about five layers at most, and the Curie constant increases with the film thickness in this range. When the film thickness is increased to 8.5 layers, Fig. 1 shows that the magnetization obeys the Curie-Weiss law. Thus we observe a crossover from the Curie to the Curie-Weiss law as the film thickness is increased.

The crossover region can be seen more clearly in Figs. 2 and 3, in which we plot parameter A and the Curie-Weiss temperature, respectively, as functions of the volume of ^{3}He condensed in the sample cell. The numbers next to the data points indicate the calculated number of layers. The error bars are the root mean square deviations of the fitted parameters. The fitted Curie constant, which is proportional to A, appears to increase linearly up to a film thickness of 3 or 4 layers. At film thicknesses greater than 5 layers, the Curie constant saturates; it remains almost unchanged between 5 and 15 layers. The solid line in Fig. 2 is the value of A calculated with the SQUID magnetometer sensitivity and assuming that the ^{3}He nuclear spins are localized and noninteracting. The measured Curie constant is consistent with the calculated line. Figure 3 shows that the fitted Curie-Weiss temperature θ exhibits crossover in the same region as the Curie constant. The fitted θ is zero from a film thickness of 1 to 4 or 5 layers, and then appears to increase relatively quickly to 0.4 mK by about 8.5 layers. This Curie-Weiss temperature is not too different from other measurements on bulk ^{3}He surrounded by other substrates.[1-4] The Curie-Weiss constant was observed when our sample cell was filled with the saturated-vapor-pressure liquid, as indicated by the square in Fig. 3.[19]

According to the density-profile calculation of the adsorbed ^{3}He film,[6] the first layer consists of highly compressed solid, the second layer is like a high-density liquid, and the additional layers behave as a saturated-vapor-pressure liquid. This simple picture of

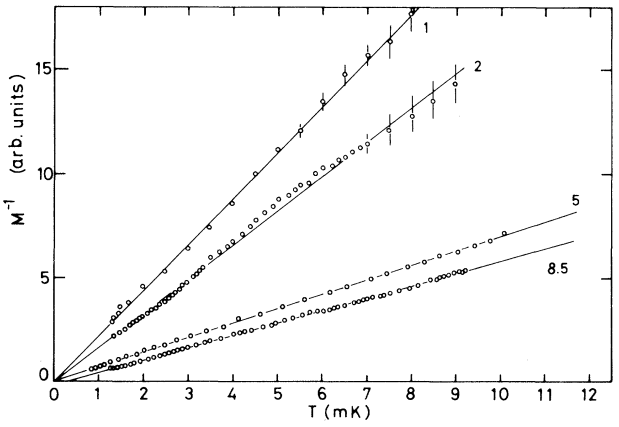

FIG. 1. Inverse of measured magnetization vs temperature for films of thickness 1, 2, 5, and 8.5 layers.

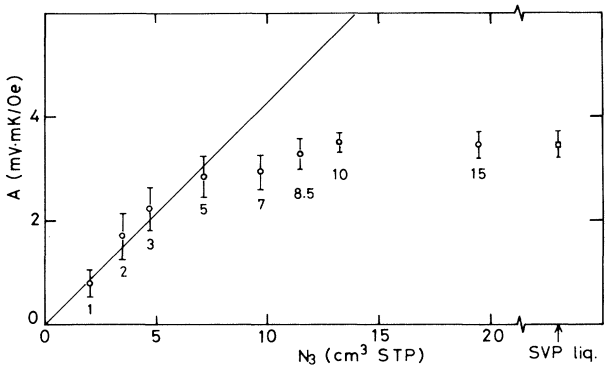

FIG. 2. Fitted parameter A (proportional to Curie constant) vs volume of adsorbed ^{3}He at STP. The numbers in the figure give the film thickness in layers. The solid line was derived, with use of our SQUID magnetometer transformer characteristics, from the Curie constant calculated on the assumption that the ^{3}He nuclear spins are free and localized.

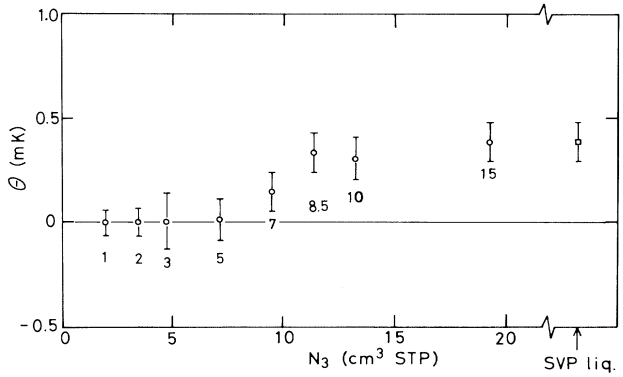

FIG. 3. Curie-Weiss temperature θ vs volume of adsorbed ^{3}He at STP.

the film implies that the magnetization of the first layer should obey the Curie law if the exchange interaction is small. Furthermore, it implies that the additional layers on top of the first layer should exhibit a temperature-independent magnetization appropriate for the degenerate Fermi liquid in the temperature range of our measurement. The above prediction for the first layer is consistent with our result. Contrary to the simple prediction, however, our results show that the magnetization continues to obey the Curie law up to a film thickness of 3 or 4 layers. It is highly unlikely that ^{3}He atoms in films as much as 4 or 5 layers thick are localized as a solid. Experiments on the superfluidity of ^{4}He films on porous substrates show that the immobile solid film is at most 1.5 layers thick.[20] The solid region for ^{3}He film would be expected to be even thinner because of the greater zero-point motion of the latter. We thus reach the surprising conclusion that the magnetization of two or three liquid layers on a first layer of ^{3}He adsorbed on silver powder exhibits the temperature dependence of a nondegenerate Fermi gas, even though the temperature range of the measurement is much lower than the Fermi temperature of bulk liquid ^{3}He.

This "induced" Curie behavior in the liquid layers on the solid layer is similar to the giant magnetic moments induced around magnetic impurities in Pd[21] or other materials[22] which exhibit a strong paramagnon effect. In the case of ^{3}He, ^{3}He nuclear spins in the liquid layer adjacent to the localized ^{3}He atoms may be affected by the polarization of the localized spins. Jichu and Kuroda[23] have claimed theoretically that large magnetic moments are induced in ^{3}He liquid next to localized ^{3}He by the exchange interaction between localized spins and those in the liquid.

The Curie constants of the films with nonzero Curie-Weiss temperatures were found to be almost the same as that for 5-layer films. This means that the spins contributing to the ferromagnetic interaction come from all 5 layers. Our result may be compared with those of Ahonen *et al.*[24] and Brewer *et al.*,[4] who claimed that the number of layers exhibiting anomalous surface magnetism is about 4. It is clear from Fig. 3 that for the ferromagnetic interaction to be initiated, it is necessary for a few more layers to be adsorbed on top of the 3–5 layers which show the Curie behavior. This result implies that the ferromagnetism observed here is closely related to the Ruderman-Kittel-Kasuya-Yosida interaction between localized ^{3}He spins and ^{3}He spins in the liquid.[24]

The magnetic behavior of ^{3}He films adsorbed on a solid substrate may be affected by magnetic impurities on the substrate. To see such an effect, the same experiment as the present one should be repeated with the substrate being coated with nonmagnetic materials such as ^{4}He or Ar. This kind of experiment is now going on and the results will be reported in a forthcoming paper.

In conclusion, the present layer-by-layer experiment on ^{3}He film adsorbed on silver powder has produced the following results:

(1) Up to a film thickness of 5 layers, the susceptibility obeys the Curie law, and the Curie constant increases linearly with the amount of ^{3}He adsorbed.

(2) When 7 to 10 layers of ^{3}He are adsorbed, the Curie constant remains unchanged and the film exhibits ferromagnetism with an interaction energy equivalent to $\theta \simeq 0.4$ mK. The number of layers contributing to the ferromagnetic interaction is about 5. The Curie constant and the ferromagnetic interaction energy remain unchanged even when the cell is filled with saturated-vapor-pressure liquid.

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[19]In a separate experiment to check for possible capillary condensation, we measured the speed of third sound on ^{4}He film on a glass substrate in good mechanical contact with sintered silver powder prepared in the same way as in the ^{3}He film experiment. The measured speed of third sound decreased with the ^{4}He film thickness up to 10 layers, in good agreement with theory, and no evidence of capillary condensation was observed, thus showing that the ^{3}He films up to 10 layers thick used in our experiment contained no capillary condensation.

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