Quantum Degeneracy and Phase Separation in Solid Two-Dimensional ^{3}He-^{4}He Mixture Films Adsorbed on Graphite

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The effects of quantum degeneracy due to the tunneling motion of ^{3}He atoms in a two-dimensional solid isotopic solution have been observed through measurements of the nuclear magnetic susceptibility. This system constitutes a solid phase with the features of a heavy fermion fluid. A phase transition, identified as isotopic phase separation, was observed directly through the time variation of the magnetic susceptibility. The critical temperature was 50 mK with a time constant for phase separation of order 5 h at 10 mK.

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The unique feature of quantum crystals is the coexistence of crystalline order with significant atomic motion, which is observable if the atoms are in some way distinguishable [1]. In pure solid ^{3}He this motion gives rise to an exchange interaction between the nuclear spins, while in sufficient dilute solutions of ^{3}He in hcp ^{4}He the excitations are delocalized Bloch-wave-like states ("mass fluctuation waves" or "impuritons") [2]. In pure solid ^{4}He it has been proposed [3] that the quantum effects can lead to a, as yet unobserved, supersolid phase. In this Letter we present evidence that in two-dimensional solid helium isotopic solutions it is possible, for the first time, to realize conditions in which the ^{3}He atoms are delocalized but at densities such that the effects of Fermi degeneracy are observable. This system therefore retains the crystalline order of a solid but simultaneously exhibits the features of a degenerate fluid of mobile heavy fermions.

The atomic tunneling rate in both bulk and two-dimensional solid helium is an extremely strong function of the lattice parameter [4]. It is an important property of helium adsorbed on graphite that both isotopes form an incommensurate two-dimensional solid with a triangular lattice [5] over essentially the same surface density range from 0.08 Å^{-2}. It is thus reasonable to hypothesize that an isotopic solution has the same structure in this density range. If so this would offer the possibility of studying two-dimensional solid mixtures at fixed surface density and structure as a function of ^{3}He concentration, and at significantly larger lattice parameters than achievable in bulk.

The potential interest of this model system can be appreciated in terms of the multiple spin exchange model [6] of atomic motion in quantum solids, according to which tunneling takes place by the cyclic permutation of rings of atoms. This model takes account of the importance of steric hindrance to exchange resulting from the hard-core repulsion between helium atoms. Semiempirical values of the exchange coefficients in bulk solid ^{3}He are in good agreement with more recent Monte Carlo calculations [7]. Since even (odd) permutations give rise to ferromagnetic (antiferromagnetic) exchange, there is competition between these various processes which reduces the observable effect of the atomic motion. Thus, for example, for bulk solid ^{3}He on the melting curve the Curie-Weiss temperature is $\theta = 2.7$ mK rather than ~ 16 mK, the value that would obtain if this cancellation did not occur [8] (i.e., "as if ^{3}He was a boson" [9]). Similarly for pure ^{3}He on a two-dimensional triangular lattice [10] $\theta = -3(J_{\rm nn} - 2J_t + 3K + 5S/8)$, where $J_{\rm nn}$, J_t, K, and S are the exchange rates for two-, three-, four-, and six-particle exchange. In contrast consider now a single ^{3}He impurity atom in a two-dimensional ^{4}He crystal of the same density. It has a hopping rate which arises from the *sum* of the contributions of all possible cyclic permutations carrying the atom to a nearest-neighbor lattice site. The tunneling (or hopping) rate is therefore [11] $t = J_{\rm nn} + 2J_t + 4K + 2S$. The search for such tunneling motion provided the motivation for these experiments.

Here we report the first measurements of the nuclear magnetic susceptibility of such a solid mixture film adsorbed on the surface of graphite between 1 K and 6 mK. The nuclear magnetic susceptibility was measured by continuous wave NMR at a frequency of 1.08 MHz using exfoliated graphite (Grafoil) as a substrate. The temperature of the film was determined using a melting curve thermometer. Measurements were made with a fixed number of adsorbed ^{3}He atoms, corresponding to 0.0132 Å^{-2}. The total coverage was varied from 0.06 to 0.14 Å^{-2} by adding ^{4}He. Samples were prepared by adding the appropriate quantity of helium gas at 4 K and annealing at 15 K. The coverage scale adopted here was that used in previous work [12].

At densities corresponding to the incommensurate solid region of the submonolayer phase diagram for the pure isotopes the measured $\chi(T)$ falls significantly below the Curie law value. This observation is consistent with the quantum degeneracy of delocalized ^{3}He atoms. However, it is complicated by a second phenomenon, namely, a phase transition within the film which we believe to be phase separation [13]. Thus if the sample is cooled from

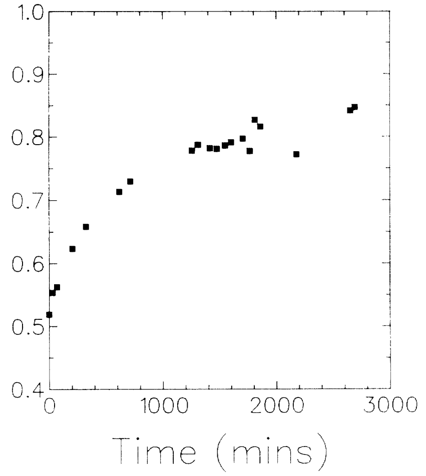

FIG. 1. Evidence for a phase transition in an isotopic mixture film (total coverage 0.082 Å^{-2}, ^{3}He concentration 0.16). Time evolution of the susceptibility on quenching to 10 mK. The product of the nuclear magnetic susceptibility and temperature is normalized by the Curie constant.

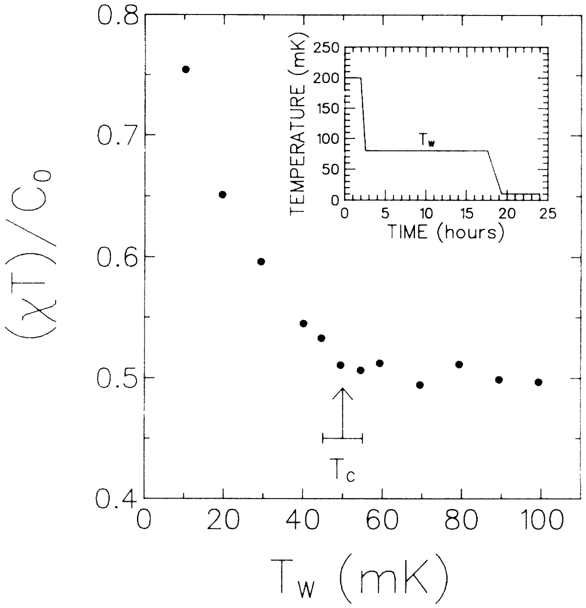

FIG. 2. Location of the critical temperature for phase separation. Inset: The temperature cycle used, where T_w is the waiting temperature (phase separation will occur here if $T_w < T_c$). The initial susceptibility at 10 mK is shown as a function of T_w.

200 to 10 mK, the observed susceptibility at 10 mK is time dependent. The initial susceptibility is significantly below the Curie law value and this we attribute to Fermi degeneracy in the homogeneous isotopic mixture. At low temperatures this phase appears unstable, we believe with respect to an isotopically phase-separated solid. The recovery in susceptibility towards the Curie law value is nonexponential and takes place over many hours [14]; the results for a film of total density 0.082 Å^{-2} are illustrated in Fig. 1. If the sample is warmed to 200 mK, held at that temperature for 2 h and subsequently cooled to 10 mK, the same behavior is observed.

It has proved possible to locate the critical temperature of this transition using the following temperature cycling procedure, illustrated in the inset to Fig. 2. The sample was warmed to 200 mK for 2 h to ensure homogeneity. It was then cooled to some intermediate temperature T_w, at which it was held for 15 h in order that phase separation might commence if $T_w < T_c$, the critical temperature. After cooling rapidly to 10 mK the initial susceptibility was determined [15]. The dependence on T_w is shown in Fig. 2 and establishes $T_c = 50 \pm 5$ mK. The characteristic time scale for the phase transition within the 2D incommensurate solid occurring when it was quenched in this way from the homogeneous state to $T < T_c$ did not appear to depend strongly on T_w. Similar behavior was found at surface densities of 0.082, 0.086, and 0.090 Å^{-2}. At higher solid densities no significant temperature dependence of χT was observed (it follows that the phase transition could not be detected by susceptibility measurements in this regime). The formation of the second layer occurs at 0.114 Å^{-2}; this density corresponds closely to that of a completed ^{4}He monolayer. ^{3}He atoms are promoted preferentially into the second layer where they form a fluid phase. Since this fluid has a Pauli susceptibility [16], while those atoms remaining in the first layer are localized and have a Curie susceptibility, this promotion can be clearly detected by measuring the reduction in susceptibility at low temperatures after each progressive addition of ^{4}He atoms.

The susceptibility of the 2D isotopic solid mixture was explored as a function of total helium coverage with the number of ^{3}He atoms held constant (corresponding to 0.0132 Å^{-2}). The susceptibility $\chi(T)$ was measured following a rapid quench from above T_c; Fig. 3 shows the temperature dependence of $\chi(T)T$ in order to emphasize deviations from Curie's law. At surface densities in the range $0.064 < \rho < 0.08$ Å^{-2} the structure of the film varies from the $\sqrt{3} \times \sqrt{3}$ commensurate phase through a partially commensurate phase or domain wall solid [4] whose structure is not firmly established. Here the departures from Curie law are relatively small and coverage independent; the peak around 25 mK has also been seen in pure ^{3}He films [12]. Large deviations from Curie law in the quenched film are seen only in the incommensurate solid and at low densities. Since in this case the film is expected to be a homogeneous isotopic mixture, this immediately suggests that here the tunneling ^{3}He atoms form a degenerate gas of two-dimensional impuritons, i.e., the ^{3}He atoms become delocalized in the regular 2D ^{4}He triangular lattice matrix. At the lower coverages, up to 0.08 Å^{-2}, the ^{3}He atoms are localized by the structural disorder [17].

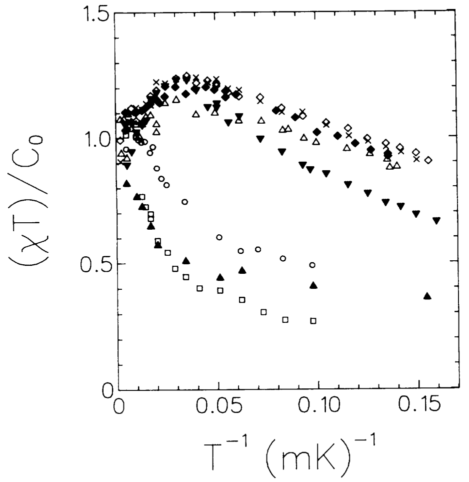

FIG. 3. $\chi(T)T$, normalized by the Curie constant, as a function of temperature, for various total coverages. The number of ^{3}He atoms is held constant. Coverages are (△) 0.064, (◇) 0.068, (◆) 0.072, (×) 0.076, (▼) 0.080, (○) 0.082, (□) 0.086, and (▲) 0.090 Å^{-2}.

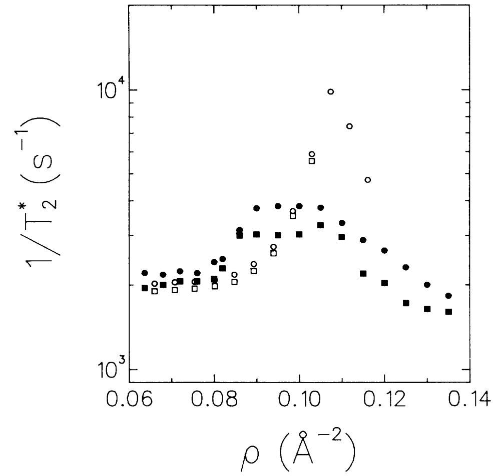

FIG. 4. Isotherms of the NMR linewidth as a function of coverage. Isotopic mixture at (■) 200 mK and (●) 10 mK; pure ^{3}He at (□) 200 mK and (○) 10 mK.

The deviations from Curie law found in the incommensurate solid are greatest at 0.086 Å^{-2}. These are consistent with a Fermi temperature of $T_F \approx 30$ mK. Using a tight-binding model [18] we find a tunneling energy $t \sim 36$ mK, corresponding to a ^{3}He effective mass $m^* \sim 20 m_3$, where m_3 is the mass of a ^{3}He atom. The large impurity atom bandwidth Δ implied by this result is much larger than the energy scale characterizing the elastic interaction between impurities [19] $\Delta > U_0$. Thus in the low surface density 2D solid the tunneling ^{3}He atoms remain weakly interacting even at the relatively large concentrations studied here. This differs markedly from ^{3}He impuritons in bulk hcp ^{4}He where $\Delta \ll U_0$ (since $\Delta \sim 0.1$ mK in the lowest density bulk hcp matrix, which has a significantly smaller interatomic separation than achievable in 2D) [1]. Thus in bulk to achieve the weakly interacting impuriton limit requires ^{3}He concentrations $x < 10^{-3}$. Furthermore, the strain interaction between impuritons results in localization [20] at $x \sim 0.1$. Thus in bulk, even without phase separation, the observation of degenerate impuritons would require extremely low concentrations and microkelvin temperatures.

The large tunneling energy we determine should be compared with the Curie-Weiss temperature in the pure ^{3}He incommensurate solid [21], where it is found that $\theta \lesssim 0.1$ mK, and NMR relaxation time measurements [3] which suggest maximum exchange energies ~ 1 mK. These results imply a tunneling rate for the hypothesized homogeneous solid mixture on a triangular lattice significantly smaller than that observed. Now it is believed that the atomic wave function normal to the surface has an important influence on the intralayer tunneling rate and that this explains the large difference between the exchange rates of the first and second layers of pure ^{3}He adsorbed on graphite [22]. Thus in the present case of the 2D isotopic solid mixture it may be that the ^{3}He atoms reside slightly further away from the substrate than the ^{4}He atoms giving rise to the enhanced tunneling rate. We note that the z component of the *single particle* atomic wave functions of both isotopes in the graphite adsorbing potential correspond closely in width and location [23]. However, there is, at solid densities, a competition between the zero point energy due to confinement in the plane and the energy associated with the motion perpendicular to the plane which would drive the proposal displacement of the ^{3}He atom. This effect should be distinguished from promotion to the second layer which occurs at the expected surface density of 0.114 Å^{-2} and has a clear experimental signature, as discussed before. On the other hand, in the phase-separated mixture each atom is surrounded by those of the same isotopic species and the slight distortion away from a strictly planar configuration of atoms would be expected to disappear. This would contribute to a reduction in tunneling rate; furthermore, in the phase-separated system the periodicity of the structure and hence the delocalized tunneling excitations are destroyed.

Measurements of the resonance linewidth, $1/T_2^*$, have a coverage dependence very different from that observed in pure ^{3}He films. In that case the linewidth increases dramatically with increasing density of the incommensurate solid, reflecting the decrease in exchange constant. A cusp indicates the decrease in correlation time following promotion of atoms to the second layer. On the other hand, the isotopic mixture data show a plateau which extends from around 0.09 Å^{-2} to second layer promotion (Fig. 4). The coverage dependence of $1/T_2^*$ is similar both above and below the phase-separation temperature. This result is corroborated by earlier data [24] taken at 1.05 K where the total coverage was varied, the number of ^{3}He atoms being held constant at an equivalent density of 0.03 Å^{-2}. This picture is not consistent with a correlation time determined by a ^{3}He-^{4}He tunneling rate that is strongly density dependent as the exchange constants in bulk and two-dimensional pure ^{3}He. However, we note that in bulk dilute isotopic mixtures the behavior of T_2 is more complex than that of the spin diffusion coefficient due to the importance of correlated motion of impuritons [25]. We are unable to detect a change in T_2 from the linewidth measurements (within 10%) during phase separation.

In conclusion, these results demonstrate for the first time the effects of Fermi degeneracy in a quantum solid and the phase separation of the two-dimensional ^{3}He-^{4}He solid mixture. The degeneracy effects occurs in the low-density incommensurate solid suggesting two-dimensional impuritons, although the magnitude of the inferred tunneling rates is not completely understood. It would be of interest to study the evolution with ^{3}He concentration from heavy fermion fluid to the exchange-dominated pure solid. Theoretically calculations of the isotopic phase-separation curve are required; here the possibility of a finite solubility of ^{3}He in 2D solid ^{4}He at low concentrations arising from quantum effects could be investigated.

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