Some Structural Properties of Solid He Films: Consequences on $^3$He Film Ferromagnetism

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Presently available experimental data on neutron diffraction, elastic properties, specific heat, high temperature susceptibility for solid helium films adsorbed on Grafoil and theoretical calculations of the energy of the fluid are used to give an estimate of first and second solid layer densities as a function of total coverage. Important information on the second layer phase diagram is deduced from this analysis. In $^3$He films, the knowledge of the second-layer-solid density $\rho_2(x)$ as a function of total coverage $x$ is essential to elucidate the origin of ferromagnetism for two solid films in the presence of a fluid third layer.

I. INTRODUCTION

The second solid layer of $^3$He adsorbed on graphite exhibits striking magnetic properties described in recent publications. In this system, a large exchange coupling between the nuclear spins is observed to evolve from antiferromagnetism to ferromagnetism as a function of coverage. Close to third layer promotion, the second layer completely solidifies. The fact that, just at the solidification the ratio of the second to the first layer density is 4/7 suggests the occurrence of a commensurate phase (although this has never been verified unambiguously by neutron diffraction). The solid second layer is characterized by an antiferromagnetic susceptibility. In the presence of a partial fluid third layer the system becomes ferromagnetic, the effect being particularly large at the “ferromagnetic peak”, close to promotion in a fourth layer. While antiferromagnetism in the low density commensurate phase has been ascribed to Multiple Spin Exchange (MSE)
effects,\textsuperscript{1,3,4} the origin of ferromagnetism at higher densities is still controvered. Multiple Spin Exchanges in the second layer are probably still efficient. As predicted in 1980 by Delrieu,\textsuperscript{5} in a high-density closed-packed lattice, the steric hinderance of hard cores favors three-particle exchange, which leads to ferromagnetism.\textsuperscript{6} However the presence of fluid above the solid films allows in principle the existence of a Ruderman–Kittel interaction which could also explain the ferromagnetism.\textsuperscript{7,8} The measurement of exchange frequencies as a function of the second layer density $p_2$ should be a crucial test: MSE frequencies vary exponentially with $p_2$, while Ruderman–Kittel interactions are expected to oscillate, as a function of the successive completion of the fluid layers. For this purpose, a precise estimate of $p_2(x)$, where $x$ is the total coverage, is needed. Previous attempts\textsuperscript{9} to relate second layer densities to the total coverage were based on an “empirical” exponential form for $p_2(x)$. Although it has the correct asymptotic behavior $p_2(x) \to \text{Cst}$ as $x \to \infty$, this functional form does not take into account the discretization corresponding to successive, third, fourth, fifth, ... fluid layer filling.

We provide here an improved estimate of $p_2(x)$ at high densities, when the second layer is an homogeneous incommensurate solid. Our calculation applies to both helium isotopes. It is based on the theoretical evaluation of the chemical potential of the fluid overlayers and experimental data on the compressibility of solid films.

A solid film ($S$) and the overlying fluid layer(s) ($L$) are first considered separately. The variation of the chemical potential of the fluid $\mu^L(p^L)$ in contact with the “substrate” (Grafoil plus one or two fixed solid helium layers) is evaluated theoretically in terms of the number of atoms in the fluid per unit area: $p^L$ (Section 2). We then estimate the chemical potential $\mu^S(p^S)$ of a solid film in terms of its density $p^S$. Since the chemical potential of a solid film is difficult to evaluate theoretically, $\mu^S(p^S)$ is deduced from experimental results on the compressibility of submonolayer films $\kappa^S(p^S)$ at various densities $p^S$, using the relation: $1/\kappa^S(p^S) = (p^S)^2 d\mu^S/dp^S$ (Section 3).

When both systems are brought together in equilibrium their respective particle numbers, $p^S$ and $p^L$, are determined by equating the chemical potentials: $\mu^S(p^S) = \mu^L(p^L)$ (Section 4). This simple procedure neglects the influence of the small variation of the solid film density $p^S$ on $\mu^L$. It also neglects the small influence of the overlying fluid on the solid film compressibility $\kappa^S(p^S)$.

The calculation is applied to an incommensurate first layer in the presence of a fluid second layer and to an incommensurate second layer in the presence of fluid third, fourth, ... layers, for both isotopes. As a test of our method, the results are compared to neutron diffraction data, when