Electron-Spin-Resonance Instability in Two-Dimensional Atomic Hydrogen Gas

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We study by electron-spin-resonance spin-polarized atomic hydrogen adsorbed on the surface of superfluid helium at temperatures T_S from 50 to 110 mK. The average dipolar field in this 2D system shifts the electron-spin-resonance peak of the adsorbed atoms relative to that of bulk atoms. The shift is directly proportional to surface density. The role of longitudinal magnetization relaxation is played by particle exchange between the 2D and the 3D phases, which diminishes exponentially with decreasing T_S. Therefore at $T_S \lesssim 80$ mK an excitation field of 0.1 mG disturbs the equilibrium surface density and leads to a magnetization instability observed as sawtooth shaped resonance lines.

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The quantum degeneracy regime in low-dimensional atomic systems is receiving increasing interest and recently progress has been made towards truly two- (2D) and one-dimensional (1D) Bose-Einstein condensates of trapped alkali vapors [1,2]. Adsorbed spin-polarized atomic hydrogen (H$\downarrow$) in the single bound state on liquid helium is a genuine 2D weakly interacting Bose gas. In the magnetic compression experiments [3], where we observed degeneracy effects in 2D H$\downarrow$, the very large field gradients used to achieve high 2D densities did not allow direct detection of atoms by magnetic resonance techniques. *In situ* detection of H$\downarrow$ by optical spectroscopy in magnetic compression experiments [4] also appears to be nontrivial. These problems may be circumvented by thermal compression of 2D H$\downarrow$ in a *uniform* field [5,6] making spin resonance applicable to direct diagnostics of the compressed gas.

Electron-spin resonance (ESR) has been successfully utilized in studies of 3D spin-polarized hydrogen. Because of a smaller number of atoms and consequent sensitivity problems the detection of 2D H$\downarrow$ is more difficult. Yet the UBC group [7,8] were able to observe by ESR that the resonance field for adsorbed H atoms is displaced from that for bulk atoms. Shinkoda and Hardy [8] found that the shift depends on the direction of the sample plane with respect to the polarizing magnetic field and attributed the shift to an internal dipolar field which, unlike in a homogeneous 3D gas, does not average to zero in 2D. The absorption line shapes of the surface atoms were very asymmetric, ramplike, and much wider than the bulk line. Motivated by the similarity between the observed line shapes and ESR lines of ferromagnetic films at very high excitation powers [9], Shinkoda and Hardy [8] considered a magnetization instability as an explanation of the spectra. However, they could not discover an effective magnetization relaxation process and thus no quantitative agreement with the experiment was obtained. Effects of internal dipolar fields are known also from NMR experiments with, e.g., liquid and solid ^{3}He [10] and with polarized ^{3}He gas in restricted geometries [11]. In these experiments a "large" spin magnetization and finite geometry of the sample, features in common with 2D H$\downarrow$, caused nonlinear demagnetization effects and consequently unusual spectral and dynamic behavior.

In this paper we describe ESR experiments where 2D spin-polarized hydrogen gas has been adsorbed on the surface of superfluid helium at temperatures from 50 to 110 mK. We find that magnetization instability effects due to high excitation microwave power are indeed the cause of asymmetric line shapes similar to those observed earlier [8]. The atom exchange between the adsorbed H$\downarrow$ and bulk gas appears to play the role of magnetization relaxation. The desorption rate decreases exponentially with decreasing surface temperature T_S and below 80 mK a microwave field as low as $H_1 \approx 0.1$ mG can be strong enough to severely distort the equilibrium density profile and consequently the magnetization, leading to the instability effects mentioned above. However, at low enough excitation levels the observed spectra do not depend on H_1 but represent a convolution of the intrinsic line shape with the spatial distribution of the internal field. The latter depends on the 2D density profile of the H$\downarrow$ sample. In the case of a homogeneous and relatively low surface density ($\sigma \approx 10^{11}$ cm^{-2}) the intrinsic linewidth is only about 20 mG.

Experiments were carried out in a 1.5 cm^3 volume sample cell (Fig. 1) located in the center of a 4.6 T superconducting solenoid. Hydrogen atoms were supplied to the cell (SC) from a 38 cm^3 buffer volume also located in the high field region and thermally isolated from the SC. This was needed to increase the lifetime of the H$\downarrow$ sample during the measurements up to about an hour. The SC and the buffer were thermally anchored to a dilution refrigerator and their temperatures were actively controlled in the respective ranges from 70 to 300 mK and 150 to 500 mK. Their inner surfaces were covered with a

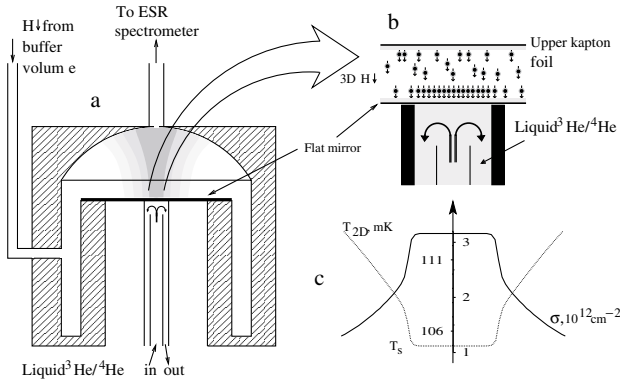

FIG. 1. Schematic drawing of (a) the sample cell with Fabry-Perot resonator, (b) flat mirror with the "cold spot," (c) calculated temperature and density profiles over the flat mirror for the ^{3}He/^{4}He coolant temperature 45 mK and the cell temperature 112 mK.

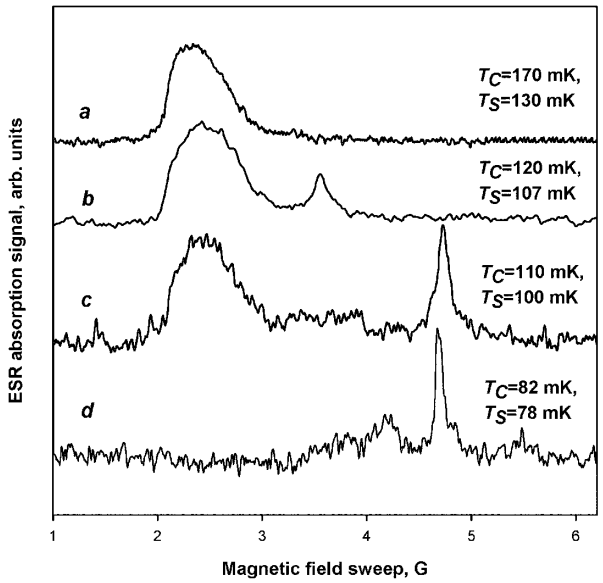

FIG. 2. Typical ESR absorption spectra in the limit of very low excitation fields taken for decreasing temperatures of the sample cell T_C and the cold spot surface T_S.

saturated film of isotopically purified ^{4}He (< 1 ppb ^{3}He). The two volumes were loaded with atomic hydrogen from a low-temperature rf dissociator.

For the detection of hydrogen in the SC by ESR there is a miniature semiconfocal Fabry-Perot resonator as a part of our 129 GHz two-phase heterodyne spectrometer. The center of the flat mirror of the resonator is thermally isolated from its outer region and is cooled to temperatures from $T_S = 30$ to 120 mK by the dilute ^{3}He/^{4}He stream of the dilution refrigerator. The flat mirror is a gold-coated 13 μm thick Kapton foil separating the coolant from the H$\downarrow$. In this way we can actively cool a 0.15 cm diameter "cold spot" (CS) independent of the rest of the SC walls. When designing the resonator we were aware of strong bulk atom signals which could mask the weak 2D lines. Therefore the volume available for the 3D gas is restricted to 1.25×10^{-3} cm^3 by another Kapton foil located 0.08 cm above the flat mirror. For our working mode TEM$_{003}$ the profile of the microwave field in the resonator is a 0.233 cm wavelength standing Gaussian wave with a waist diameter of 0.31 cm at the flat mirror. We are able to detect down to 10^9 hydrogen atoms with an excitation power of 0.1 nW incident on the resonator. Low powers are required to minimize the ESR-induced destruction of the sample. The absolute values of the bulk density n are obtained with an accuracy of 20% by integrating the bulk absorption lines. The latter are calibrated calorimetrically against the total recombination heat released during the decay of the sample.

Figure 2 shows typical ESR absorption spectra recorded in the limit of very low excitation fields for the $b \rightarrow c$ hyperfine transition of atomic hydrogen. The bulk gas signal [trace (a)] can be easily detected at relatively high wall temperatures, when the surface density σ is still very low. The signal is inhomogeneously broadened and asymmetric, representing a convolution of the intrinsic line shape with the profiles of the main magnetic field and the microwave field in the resonator. At lower T_S an extra peak appears at a higher field than the bulk gas line [trace (b)]. It is associated with the 2D atoms and, when T_S decreases further and σ increases, it moves farther away from the bulk line [trace c)]. Ultimately the bulk density becomes so small that the 3D resonance vanishes in the noise and only the 2D line remains [trace (d)].

In the present range of temperatures and densities the bulk gas is in dynamic equilibrium with the adsorbed phase due to a balance between the adsorption and desorption rates. Then the surface and bulk densities are, in the classical limit, related through the adsorption isotherm

$$\sigma = n\Lambda \exp(E_b/k_B T_S), \tag{1}$$

where Λ is the thermal de Broglie wavelength and $E_b/k_B = 1.14$ K [12] is the binding energy of hydrogen to liquid ^{4}He. For $T_S < 100$ mK the exponential factor in Eq. (1) is very large and even a relatively small decrease in T_S leads to a large increase of σ on the CS as compared with the rest of the SC walls at a temperature $T_C > T_S$. The surface density profile over the flat mirror is determined by the temperature profile of the helium substrate. Figure 1(c) shows an example of the calculated T_S and σ profiles [13].

The width of the observed surface line was found to depend on the temperature difference $T_C - T_S$. A large difference implies a broad σ profile and hence wider lines. The situation $T_C - T_S = 0$ is of special interest because it allows one to observe the intrinsic line shape of the adsorbed atoms and to define the line position better. We made experiments where T_S and T_C were set equal in the

range 90 to 120 mK. Then the narrowest linewidths recorded for $\sigma = 2 \times 10^{11}$ cm^{-2} were about 20 mG widening to 200 mG for $\sigma = 2 \times 10^{12}$ cm^{-2}. Because of the relatively low signal-to-noise ratio we were unable to perform a quantitative study of the shape of the 2D lines at very low powers. Yet, the line shifts ΔH from the bulk resonance were obtained with an accuracy of 10 mG. Figure 3 shows the results of such measurements. At low σ the recombination overheating of the CS can be neglected and one may use Eq. (1) to calculate σ from the measured n. On the basis of the data of Fig. 3 we conclude that the displacement of the ESR line of adsorbed atoms from the bulk line is linearly proportional to the surface density such that $\Delta H = A\sigma$, where $A = 0.9(3) \times 10^{-12}$ G cm^2.

The influence of the excitation power on the 2D line shapes is demonstrated in Fig. 4. At a high power level the low-field side of the ESR response becomes an abrupt jump which with increasing power moves towards the bulk line. In an upward sweep the jump takes place in a higher field than in a downward sweep [trace (*d*)]. An order of magnitude increase in the excitation level results in very broad lines resembling those observed at UBC [8]. These effects can be explained by considering the balance between adsorption of atoms from the bulk gas and the loss of atoms from the 2D phase due to desorption and ESR-induced recombination. The adsorption and desorption rates are, respectively, $n\upsilon s/4$ and σ/τ_s, where s is the sticking probability, υ is the thermal speed of the bulk atoms, and the surface residency time is given by $\tau_s = (4\Lambda/\upsilon s)\exp(E_b/k_B T_S)$. For surface densities of interest, $\sigma > 10^{11}$ cm^{-2}, the characteristic recombination time of a spin-flipped atom is $\tau_{bc} \lesssim 3$ μs [12]. On the other hand, at temperatures below 110 mK, $\tau_s \gtrsim 100$ μs $\gg \tau_{bc}$, and therefore each spin flip instantly removes two particles from the 2D sample. Thus, neglecting spontaneous recombination processes of H$\downarrow$, a stationary surface density during the ESR sweep can be found from the particle balance equation

$$\frac{n\upsilon s}{4} - \frac{\sigma}{\tau_s} - 2\sigma\frac{\pi}{8}\gamma_e H_1^2 f(h - A\sigma) = 0, \qquad (2)$$

where H_1 is the amplitude of the linearly polarized mm-wave field in the sample, γ_e is the electron gyromagnetic ratio, $f(h - A\sigma)$ is the normalized intrinsic line shape function, and h is the sweep field relative to the bulk resonance. The adsorption/desorption exchange of atoms obviously plays the role of an effective T_1 relaxation process in 2D H$\downarrow$. Equation (2) holds also for a spatially inhomogeneous case if the in-plane transport of particles is insignificant. Then $f(h)$ represents an inhomogeneously broadened line form. Trace (*e*) in Fig. 4 is a line shape calculated from Eq. (2).

For our relatively slow sweeps through the resonance the stationary density and thus the line shift decrease with growing H_1. Such a behavior is like a typical saturation effect of an ESR response. It results in a "pulling" of the line Δh_p towards the bulk resonance given now by

$$\Delta h_p \approx \frac{\pi}{4}\tau_s\gamma_e H_1^2 f(0) A\sigma \approx \frac{\pi}{4}\gamma_e H_1^2 \tau_s \frac{A\sigma}{\Gamma}. \qquad (3)$$

Here Γ is the effective half-width of the function $f(h)$. From the measurements we found that the line pulling

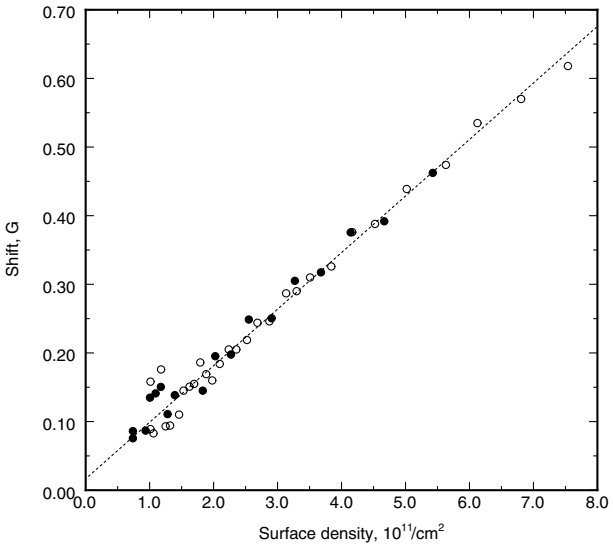

FIG. 3. Shift of the 2D hydrogen ESR line from the bulk resonance. (•) $T_C = T_S = 112$ mK; (∘) $T_C = T_S = 103$ mK. The coinciding slopes for different temperatures indicate that a correct value of E_b is used.

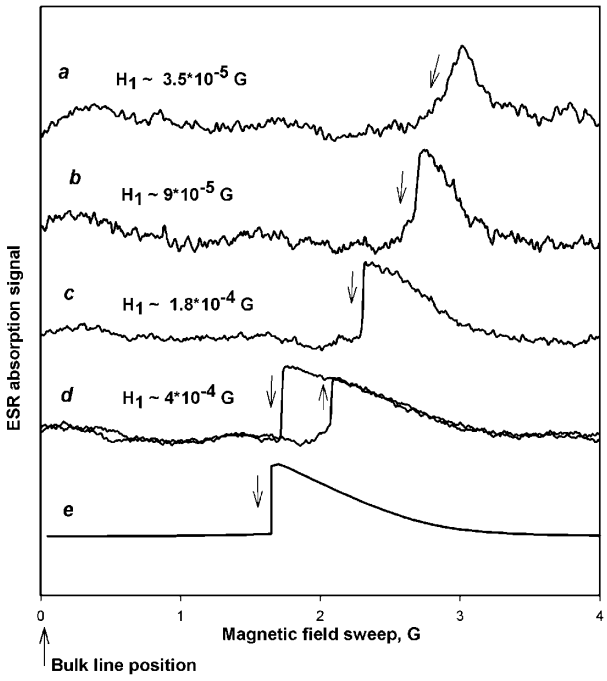

FIG. 4. ESR spectra of 2D H$\downarrow$ for various excitation fields H_1 and for $n = 2 \times 10^{13}$ cm^{-3}, $T_C = 112$ mK, and $T_S = 92$ mK. Curve (*e*) is the solution of Eq. (2) for $H_1 = 5.5 \times 10^{-4}$ G.

saturates with increasing excitation power and is essentially independent of density. Since the desorption rate σ/τ_s grows rapidly with increasing surface temperature, the former effect can be explained by recombination heating increasing with the increasing flipping rate. The latter observation may be attributed to collisional broadening which makes the intrinsic linewidth Γ proportional to σ.

For a given h Eq. (2) may have several different solutions for σ which are responsible for the observed hysteresis of the ESR spectra. Physically the instability is a positive feedback of the system's response to microwave excitation: Approaching the wing of the resonance curve from the low-field side decreases the stationary density and moves the position on the spectrum closer to the resonance maximum. This happens when the line pulling exceeds Γ and then, omitting insignificant constants, we may write the instability condition in the form

$$H_1^2 \gtrsim \Gamma^2/\gamma_e \tau_s A\sigma. \tag{4}$$

When applied to our case, the criterion for instability in ferromagnetic films [9] reads

$$H_1^2 \geq C\Gamma^3/A\sigma, \tag{5}$$

where $C = 3.08$ for the Lorentzian line form. It should be pointed out that the occurrence of instability does not presuppose anything about the origin of the surface line shift as long as the latter is proportional to the surface density.

Condition (5) assumes equal longitudinal and transverse relaxation times, $T_1 = T_2 = 1/\gamma_e\Gamma$, which is clearly not the case for 2D H$\downarrow$. Following the arguments of Shinkoda and Hardy [8] who considered a collisional origin of the intrinsic linewidth, we get at the estimate $\Gamma \approx 2A\sigma(\mu\gamma_e/va^2) \approx 0.05A\sigma$, where $a = 0.37$ nm is the radius of nearest approach. At a typical density of $10^{12}\,\mathrm{cm}^{-2}$ this corresponds to $T_2 \approx 1\ \mu\mathrm{s} \ll T_1$. Obviously the use of (5) instead of (4) leads to a greatly overestimated critical excitation field H_1 [14].

Because of the strong temperature dependence of the surface residency time τ_s condition (4) can be easily fulfilled at sufficiently low temperatures even for relatively low excitation fields. Undoubtedly the instability was also responsible for the unusual line shapes observed by Shinkoda and Hardy [8]. Therefore in their case the 2D line shift was not measured quite correctly and the area under the absorption line could not be used to extract the surface density. To avoid the instability one has to keep the excitation power low enough and to scale it as $\exp(-E_b/T_s)$. This makes studies of undisturbed 2D line shapes at temperatures well below 100 mK difficult. On the other hand, performing such experiments deeper in the quantum degenerate regime might offer a possibility to detect superfluidity of adsorbed H$\downarrow$ through its influence on the 2D hydrodynamics and consequently on the inhomogeneous broadening of the 2D ESR line. The highest 2D phase-space density we have reached in these experiments by thermal compression is 1.5.

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[1] A. Görlitz *et al.*, Phys. Rev. Lett. **87**, 130402 (2001).
[2] S. Burger, F. S. Cataliotti, C. Fort, P. Maddaloni, F. Minardi, and M. Inguscio, Europhys. Lett. **57**, 1 (2002).
[3] A. I. Safonov, S. A. Vasilyev, I. S. Yasnikov, I. I. Lukashevich, and S. Jaakkola, Phys. Rev. Lett. **81**, 4545 (1998).
[4] A. P. Mosk, M. W. Reynolds, T. W. Hijmans, and J. T. M. Walraven, Phys. Rev. Lett. **81**, 4440 (1998).
[5] B. V. Svistunov, T. W. Hijmans, G. V. Shlyapnikov, and J. T. M. Walraven, Phys. Rev. B **43**, 13 412 (1991).
[6] A. Matsubara, T. Arai, S. Hotta, J. S. Korhonen, T. Suzuki, A. Masaike, J. T. M. Walraven, and T. Mizusaki, Physica (Amsterdam) **194B–196B**, 899 (1994).
[7] M. W. Reynolds, I. Shinkoda, W. N. Hardy, A. J. Berlinsky, F. Bridges, and B. W. Statt, Phys. Rev. B **31**, 7503 (1985).
[8] I. Shinkoda and W. N. Hardy, J. Low Temp. Phys. **85**, 99 (1991).
[9] P. W. Anderson and H. Suhl, Phys. Rev. **100**, 1788 (1955), and references therein.
[10] J. Jeener, Phys. Rev. Lett. **82**, 1772 (1999), and references therein.
[11] E. Stoltz, J. Tannenhauser, and P. J. Nacher, J. Low Temp. Phys. **101**, 839 (1995).
[12] A. I. Safonov, S. A. Vasilyev, A. A. Kharitonov, S. T. Boldarev, I. I. Lukashevich, and S. Jaakkola, Phys. Rev. Lett. **86**, 3356 (2001).
[13] A detailed model accounting for various recombination and relaxation processes and hydrodynamic flow of H$\downarrow$ on the surface will be described elsewhere.
[14] I. Shinkoda, Ph.D. thesis, University of British Columbia, 1990.