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**Citation for published version (APA):**

Koelman, J. M. V. A., Noteborn, H. J. M. F., Goey, de, L. P. H., Verhaar, B. J., & Walraven, J. T. M. (1985). Spin waves in spin-polarized hydrogen atoms adsorbed on a superfluid 4He film. *Physical Review B: Condensed Matter*, 32(11), 7195-7199. <https://doi.org/10.1103/PhysRevB.32.7195>

**DOI:**

[10.1103/PhysRevB.32.7195](https://doi.org/10.1103/PhysRevB.32.7195)

**Document status and date:**

Published: 01/01/1985

**Document Version:**

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

**Please check the document version of this publication:**

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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## Spin waves in H $\downarrow$ adsorbed on a superfluid $^4\text{He}$ film

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(Received 16 July 1985)

The possibilities for observation and the properties are discussed for two-dimensional spin waves in a nondegenerate spin-polarized atomic hydrogen gas adsorbed on a superfluid helium film. We present results for the spin-transport parameters  $D_0$  and  $\mu$ , based on two-dimensional effective-range theory. The spin-wave quality factor is an order of magnitude smaller than in the volume case.

### I. INTRODUCTION

Spin waves in ferromagnets and other dense systems have been known since the 1950s. In such systems the de Broglie wavelength of the constituents is at least comparable to the distance between nearest neighbors. The associated quantum exchange interaction generated by identical-particle symmetrization is known to play a crucial role in the propagation of spin waves. Some years ago Bashkin<sup>1</sup> and Lhuillier and Laloë<sup>2</sup> pointed to the less obvious possibility of spin waves in very dilute nondegenerate gases. The existence of spin waves was demonstrated a year ago in spin-polarized atomic hydrogen<sup>3,4</sup> and at about the same time<sup>5</sup> in spin-polarized  $^3\text{He}$ . This discovery led us to investigate the possibility of such waves propagating in the two-dimensional H $\downarrow$  gas adsorbed on a superfluid helium film.

Such surface spin waves would be interesting for their own sake, but also from a more general point of view. Taking into account the important role of surface atoms in the decay of H $\downarrow$  in stabilization experiments, it is of vital importance to confirm the accepted picture that the collisions of adsorbed H atoms are not influenced significantly by the dynamics of the helium film. By now it becomes clear<sup>6</sup> that the decay of the atomic density at the surface is not primarily due to two-body dipolar relaxation and thus the latter process does not produce the useful information on the above-mentioned properties which would otherwise have been obtained. The three-body surface collision processes which one now tends to hold responsible for the decay are probably too complicated to yield such reliable information. On the contrary, surface properties derived from one- and two-body processes such as spin waves are indispensable for reliable calculations<sup>7</sup> of three-body decay at the surface. When it is possible to measure specific transport coefficients such as  $D_0$  and  $\mu$  (see below) for surface spin waves, this would yield valuable information on the H $\downarrow$ +H $\downarrow$  surface scattering. A preliminary report on the present work was presented in Ref. 8.

Previous detailed enquiries<sup>9</sup> into the extent of three dimensionality of the H-H collision process at a super-

fluid  $^4\text{He}$  surface, as well as the development of a two-dimensional effective-range theory,<sup>10</sup> provide us with sufficient insight to calculate the properties of spin waves in adsorbed H $\downarrow$  assuming a static  $^4\text{He}$  surface. Recently, Bashkin<sup>11</sup> also discussed the possibilities for observation of spin waves in adsorbed H $\downarrow$ . He used a scaling procedure to relate the two-body surface scattering process to that in three dimensions. The premisses for applying this scaling transformation are certainly not fulfilled for H $\downarrow$  on  $^4\text{He}$ , in which we are primarily interested in this paper in view of the experience from Ref. 9: the width  $d$  of the atomic wave functions perpendicular to the surface is of the order of the range of the H-H triplet interaction. If one would nevertheless apply it to that case, the result for the spin wave quality factor is of the order of  $d/a$  independent of temperature, where  $a$  is the three-dimensional (3D) scattering length. This value is a factor of 5 larger than that to be obtained in the following from a more reliable approach, a factor which may be of crucial importance in connection with the prospects for observation of surface spin waves on  $^4\text{He}$ . Contrary to Bashkin we shall also pay attention to the consequences of the adsorption-desorption kinetics for the observability of surface spin waves.

### II. SURFACE SPIN WAVES

On the atomic scale H $\downarrow$  spin waves are due to the "identical spin rotation" (ISR) effect: In the case of complete polarization and small tipping angles the effective spins precess in a two-body collision about their sum over an angle  $-2\epsilon\delta(k)$ , where  $\epsilon=+1$  ( $-1$ ) for bosons (fermions) and  $\delta(k)$  is the  $s$ -wave phase shift calculated for a spin-independent potential. Low temperatures are essential for the ISR to lead to a coherent spin transport through the medium. On one hand to avoid collisions with  $k$  values for which  $|2\delta(k)|=O(1)$ . On the other hand, to avoid  $p$  and higher waves which also perturb the simple ISR behavior.

On the macroscopic scale the collective spin dynamics is described by spin-wave equations in which two important parameters are  $D_0$ , the spin-diffusion constant in the

unpolarized gas, and  $\mu$ , measuring the influence of the particle indistinguishability on the spin transport properties. In  $d$  dimensions we have for a nondegenerate spin-polarized gas,

$$n_d D_0 = \frac{2\pi d \hbar}{2^{d/2} m \lambda^d} \left\{ \int_0^\infty d\xi e^{-\xi^2} (2\xi)^3 \sin^2 \left[ \delta \left( \frac{\sqrt{2\pi}}{\lambda} \xi \right) \right] \right\}^{-1}, \quad (1)$$

$$\mu = \frac{\int_0^\infty d\xi e^{-\xi^2} \xi^3 \cos \left[ \delta \left( \frac{\sqrt{2\pi}}{\lambda} \xi \right) \right] \sin \left[ \delta \left( \frac{\sqrt{2\pi}}{\lambda} \xi \right) \right]}{\int_0^\infty d\xi e^{-\xi^2} \xi^3 \sin^2 \left[ \delta \left( \frac{\sqrt{2\pi}}{\lambda} \xi \right) \right]}, \quad (2)$$

where  $\lambda$  is the thermal wavelength and  $n_d$  is the  $d$ -dimensional particle density. Turning to  $d=2$  we use the effective-range expression<sup>10</sup>

$$\cot[\delta(k)] = \frac{2}{\pi} [\gamma + \ln(\frac{1}{2}ka)] + \frac{1}{2\pi} r_e^2 k^2, \quad (3)$$

with  $\gamma = 0.57721 \dots =$  Euler's constant, while  $a = 2.3a_0$  and  $r_e = 14.3a_0$ , the two-dimensional scattering length and effective range, respectively. These values were calculated using a potential obtained by averaging the H-H triplet potential over the finite extension of the atomic wave functions perpendicular to the surface (so-called  $2\frac{1}{2}$ -D model<sup>9</sup>). Figure 1 shows  $n_2 D_0$  and  $\mu$  as functions of temperature. For low temperatures ( $T < 0.2$  K) these coefficients go to the values

$$\mu \simeq (2/\pi) [\gamma + \ln(3\pi a / 4\sqrt{2}\lambda)]$$

and

$$n_2 D_0 \simeq (\pi \hbar / 2m \lambda^2) / \sin^2(\operatorname{arccot} |\mu|).$$

The value for  $\mu$ , as well as the corresponding spin-wave quality factor, following from the first of these equations, is considerably smaller than the bulk value. Note, furthermore, that  $\mu$  shows a weak temperature dependence, which is due to the typical logarithmic  $k$  dependence of the phase shift in two dimensions.

To investigate under what conditions H $\downarrow$  spin waves might be observable in the adsorbed phase we consider a Cornell-type NMR experiment using a cell with a large surface to volume ratio and most of the surface parallel to the (linear) magnetic field gradient in the  $x$  direction. Of the remaining small surface part one end is at  $x=0$  and the other at  $x=L$ . Following Ref. 4 we denote the component of the polarization along  $\mathbf{B}$  by  $\sigma_0$  and its positive circular component in the frame rotating with the Larmor frequency at  $x=0$  by  $\delta\sigma_+$ . For the geometry considered we are interested in the lowest transverse ( $yz$ -independent) mode, being the only transverse mode coupled to the NMR resonator. We thus have  $\delta\sigma_+(\mathbf{r}, t) = F(x, t)$ , where  $F$  satisfies the boundary condition, based on the smallness of the end surfaces,

$$\frac{\partial F}{\partial x} \Big|_{x=0} = \frac{\partial F}{\partial x} \Big|_{x=L} = 0, \quad (4)$$

and the mode expansion

$$F(x, t) = \sum_k F^k(x) e^{-i\omega_k t}, \quad (5)$$

in which  $F^k(x)$  satisfies

$$\frac{d^2 F^k}{dx^2} = \frac{\epsilon\mu\sigma_0 - i}{D_0} \left[ \omega_k - \frac{Gx}{\hbar} \right] F^k, \quad (6)$$

and the boundary conditions (4). In Eq. (6),  $Gx/\hbar$  is the shift of the Larmor frequency due to the field gradient.

Without  $\epsilon\mu\sigma_0$  term Eq. (6) represents spin diffusion in an inhomogeneous field. The  $\epsilon\mu\sigma_0$  term arises from the ISR effect. Its form can be understood qualitatively by considering the net effect on a spin of competing ISR precessions around neighboring spins. Clearly, this net effect vanishes for spatially constant or linearly varying polarization  $\sigma(x, t)$ . The first nonvanishing contribution comes from the second derivative. This can be visualized by studying the behavior of a single spin due to two neighboring spins. The net ISR effect vanishes when the latter are tilted over the same angle in opposite directions relative to the first one. Only deviations from this situation contribute. The corresponding molecular field term in the equation for  $\delta\sigma/\delta t$  gives rise to the above-mentioned  $\epsilon\mu\sigma_0$  term in Eq. (6).

For frequencies  $\omega_k$  small relative to  $L |G|/\hbar$  the solutions of Eqs. (6) and (4) can be expressed<sup>12</sup> in Airy functions  $\text{Ai}$ , each of which corresponds to a (complex) spin-wave eigenfrequency,

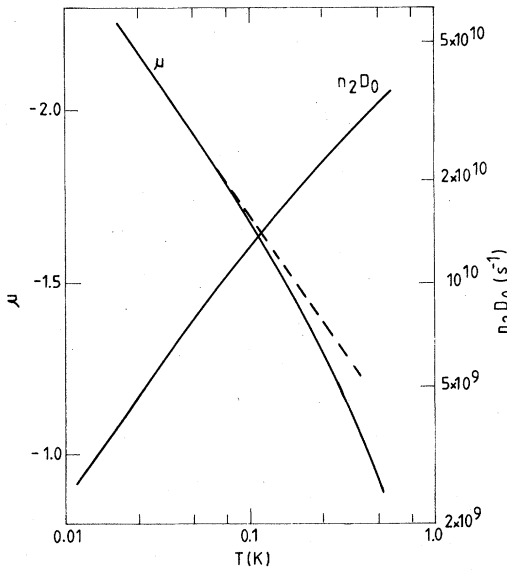


FIG. 1. Two-dimensional spin-transport coefficients  $n_2 D_0$  and  $\mu$  versus temperature. The broken line indicates the low-temperature limit for  $\mu$  which depends logarithmically on temperature.

$$\omega_k = \pm a_k \left( \frac{D_0 G^2 / \hbar^2}{|\mu \sigma_0| \mp i} \right)^{1/3}, \quad (7)$$

with  $a_k$  defined as the  $k$ th zero of  $\text{Ai}'$ , being negative real for all values  $k=1, 2, \dots$ . In Eq. (7) and in the following the upper sign refers to the case  $\epsilon \mu \sigma_0 > 0$  and the lower sign to  $\epsilon \mu \sigma_0 < 0$ . Writing  $\omega_k$  as  $\Omega_k - i\Gamma_k$ ,  $\Omega_k$  and  $\Gamma_k$  are products of  $a_k$  and  $k$ -independent quantities:

$$\Omega_k = \pm a_k \Lambda \cos \theta, \quad \Gamma_k = -a_k \Lambda \sin \theta. \quad (8)$$

The distance between eigenfrequencies is determined by the constant

$$\Lambda = \left( \frac{D_0^2 G^4 / \hbar^4}{\mu^2 \sigma_0^2 + 1} \right)^{1/6}, \quad (9)$$

while the quality factor  $Q = |\Omega_k| / \Gamma_k = \cot \theta$  is determined by the constant

$$\theta = \frac{1}{3} \text{arccot} |\mu \sigma_0|. \quad (10)$$

The observed spectrum associated with Eq. (5) is a sum over Lorentz profiles centered at the frequencies  $\Omega_k$  with half-width  $\Gamma_k$ . Notice that  $\omega=0$  corresponds to the highest (lowest) Larmor frequency in the sample in case  $\epsilon \mu \sigma_0 > 0$  ( $< 0$ ). Hence for negative polarization ( $\sigma_0 < 0$ ) and repulsive ( $\mu < 0$ ) bosons ( $\epsilon = +1$ ) the spin-wave frequencies  $\Omega_k$  of the most weakly damped spin waves add positively to the mean Larmor frequency, so that the sharpest spin-wave peaks appear on the high-frequency side of the resonance spectrum.

### III. ADSORPTION-DESORPTION KINETICS

In the foregoing analysis we assumed the surface spin transport to be decoupled from that in the bulk. In what circumstances does the adsorption-desorption kinetics allow for such a decoupling? We consider the following two time scales:  $\tau_d = 1/\Gamma_1$ , i.e., the damping time of the most weakly damped surface wave, and the mean residency time of atoms on the surface:

$$\tau_{\text{res}} = \frac{\pi m_H \lambda^2}{8 \hbar \alpha} e^{E_B / k_B T}. \quad (11)$$

We take<sup>13</sup> the sticking probability  $\alpha$  equal to 0.03 and the binding energy<sup>14</sup> in temperature units  $E_B / k_B$  equal to 1.0 K.

The adsorption-desorption kinetics does not influence the surface spin-wave phenomenon if

$$\tau_{\text{res}} \gg \tau_d. \quad (12)$$

We define the auxiliary time constant  $\tau' = (n_2)^{1/2} \hbar / |G|$ . This is the time which would be needed by two H atoms at the average interparticle distance to undergo a relative spin precession of 1 rad. For typical densities  $n_2 \approx 10^9 \text{ cm}^{-2}$  and field gradients  $|\nabla B| \approx 10^{-4} \text{ T cm}^{-1}$ ,  $\tau'$  is of the order of 1 s.

For complete polarization we then have

$$\tau_d = \frac{-(\mu^2 + 1)^{1/6} (\tau')^{2/3}}{a_1 (n_2 D_0)^{1/3} \sin(\frac{1}{3} \text{arccot} |\mu|)}. \quad (13)$$

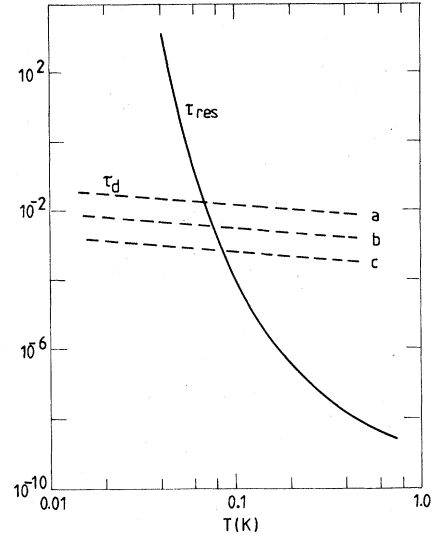


FIG. 2. The times  $\tau_d$  and  $\tau_{\text{res}}$  as a function of temperature (vertical scale in seconds) for  $\tau' = 10$  s (curve a), 1 s (curve b), and 0.1 s (curve c).

Figure 2 shows  $\tau_d$  for various values of  $\tau'$ , as well as  $\tau_{\text{res}}$ , as a function of temperature. Clearly, for the above-mentioned  $n_2$  and  $|\nabla B|$  values condition (12) is fulfilled for temperatures below 0.08 K. Considering from now on this regime, the spin-wave peaks have a typical width  $\tau_d^{-1} \approx 10^2 \text{ s}^{-1}$ . This value is comparable to that for bulk spin waves, as is the total width  $L |G| / \hbar \approx 10^4 \text{ s}^{-1}$  (dimension  $L \approx 1 \text{ cm}$ ) of the NMR absorption spectrum. Due to the lower  $|\mu|$  value, however, the quality factor is an order of magnitude smaller than for bulk spin waves. Thus, surface spin-wave peaks in the spectrum are as narrow as in the volume case, but their mutual distance is smaller.

From the point of view of observability it is also of importance to point out that the surface resonance spectrum is shifted over  $2.5 \times 10^4 \text{ Hz}$  by the surface hyperfine frequency shift relative to the volume spectrum. For the overall intensity of the surface signal the total number of atoms is of interest. It is larger or comparable to the number of volume atoms for surface to volume ratios  $A/V > 7 \text{ cm}^{-1}$  ( $T \approx 0.08 \text{ K}$ ). With a high-field ( $\approx 8 \text{ T}$ ) NMR spectrometer the minimum detectable number<sup>15</sup> of H atoms is of order  $3 \times 10^{13}$ . For detection of spin waves using small tipping angles, substantially larger quantities are required. This implies the necessity of large surface area within the resonator, possibly a large number of sheets or a ribbon. It seems questionable whether sufficient surface area may be realized in practice. In principle, a large gain in sensitivity may be realized by working at  $B = 0.65 \text{ T}$ , where the NMR frequency is field independent to first order.<sup>16</sup> Since this implies a different excitation and detection scheme it is not further discussed here. We also have to take into account the requirement that  $\tau_d$  should be small relative to the recombination time for H atoms at the surface. The experimental value<sup>6</sup> for  $L_s$  indicates, however, that this requirement is amply fulfilled.

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