

# Influence of Film-Mediated Interactions on the Microwave and Radio Frequency Spectrum of Spin-Polarized Hydrogen on Helium Films

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We argue that helium film-mediated hydrogen-hydrogen interactions strongly reduce the magnitude of cold collision shifts in spin-polarized hydrogen adsorbed on a helium film. With plausible assumptions about experimental parameters this can explain (i) the 2 orders of magnitude discrepancy between previous theory and recent experiments and (ii) the anomalous dependence of the cold collision frequency shifts on the film's  $^3\text{He}$  covering. The mediated interaction is attractive, suggesting that in current experiments the gas will become unstable before reaching the Kosterlitz-Thouless transition.

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Two-dimensional (2D) dilute, spin-polarized hydrogen is a quantum fluid that is actively studied by both the cold atom and low temperature communities. One motivation for this interest is the belief that 2D hydrogen can support a superfluid-normal Kosterlitz-Thouless transition, which crosses over to Bose-Einstein condensation (BEC) as the confinement length in the third-dimension is increased. The relatively large particle density and light mass give a high BEC temperature. Since the hydrogen-hydrogen interaction potential is accurately known, it is widely believed that one will be able to quantitatively compare theory and experiment.

Despite this quantitative understanding of dilute hydrogen, experiments on spin-polarized hydrogen adsorbed on a helium film display a cold collision frequency shift that is 2 orders of magnitude smaller than theory predicts [1]. This suggests that the helium, though traditionally considered inert, significantly affects the hydrogen. Here we show that a helium mediated interaction alters the hydrogen collision properties and resultantly reduces the cold collision shift. The exact magnitude of this shift depends on experimental parameters such as the helium film thickness. For some reasonable estimates of these parameters, we find that the magnitude of this reduction is sufficient to eliminate the discrepancy. Using these same estimates, we find a counterintuitive dependence of the shift on the  $^3\text{He}$  concentration, just as seen in the experiments. We do not, however, find quantitative agreement with the shift's magnitude.

As elaborated below, Ahokas *et al.* [1] measure a two-photon radio frequency (rf)-microwave absorption spectrum. These experiments are in the “cold collision” regime [2], where the thermal de Broglie wavelength is longer than the interaction's effective range, resulting in extremely sharp spectral lines, which are shifted by interactions. These shifts are important: they limit the performance of technologies (hydrogen masers [3,4] and atomic clocks [5]) and are versatile probes. For example, related spectral shifts were used to diagnose BEC in 3D spin-polarized

hydrogen [6], identify Mott insulating shells in optical lattices [7], and reveal pair structure in Fermi gases [8,9]. Future experiments will rely on these techniques to explore ever more exotic phenomena.

*Experimental details.*—Ahokas *et al.* [1] study a 2D hydrogen gas bound by van der Waals forces at a distance  $\zeta \sim 5 \text{ \AA}$  above a helium film, working in a uniform magnetic field  $B = 4.6 \text{ T}$ . They produce a cloud with more than 99% of the atoms initially in the lowest hyperfine state  $|1\rangle$ . Letting  $|s_e s_n\rangle$  be the state with electron spin  $s_e$  and nuclear spin  $s_n$ , one finds  $|1\rangle = \cos\theta|\uparrow\uparrow\rangle - \sin\theta|\uparrow\downarrow\rangle$ , with  $\theta = 5.5 \times 10^{-3}$ . Driving a transition to state  $|2\rangle = \cos\theta|\uparrow\downarrow\rangle + \sin\theta|\downarrow\uparrow\rangle$ , the experimentalists measure spectra by counting the number of transferred atoms for a given probe detuning. They vary temperature, 2D hydrogen density, and the He film's  $^3\text{He}$  concentration from negligible to  $10^{14} \text{ cm}^{-2}$ .

*Calculating spectra.*—Neglecting, for now, the helium film's degrees of freedom, the hydrogen's Hamiltonian is

$$H = \sum_{j,k} \epsilon_{j,k} \psi_{j,k}^\dagger \psi_{j,k} + \sum_{\substack{i,j \\ k,p,q}} \frac{V_{ij,q}}{2} \psi_{j,p}^\dagger \psi_{i,k}^\dagger \psi_{i,k+q} \psi_{j,p-q}, \quad (1)$$

where  $\psi_{j,k}^\dagger$  are bosonic creation operators for momentum  $\hbar k$  and internal state  $j$ ;  $\epsilon_{j,k} = \hbar^2 k^2 / 2m + \delta_j - \mu_j$  is the free dispersion relation of the effectively 2D, mass  $m$  hydrogen, including the internal energy  $\delta_j$  and chemical potential  $\mu_j$ ;  $V_{ij,k}$  is the Fourier space interaction potential between atoms in states  $i$  and  $j$ ; and  $\hbar$  is the reduced Planck's constant.

In the rotating wave approximation the probe's Hamiltonian [10] is  $H_P = \Omega_P \sum_k e^{-i(\omega - (\mu_2 - \mu_1))t} \psi_{2,k}^\dagger \psi_{1,k} + \text{H.c.}$  The rf and microwave photons transfer negligible momentum. Given that the range of the potential ( $\sim 1 \text{ \AA}$ ) is significantly less than the 2D interparticle separation ( $\sim 100 \text{ \AA}$ ), the rf or microwave spectrum remains a delta function [2], with a shift given by [11]

$$\delta\omega = \frac{\hbar^2}{m} g_2(0) [f_{12}(q_T) - f_{11}(q_T)] \sigma_1, \quad (2)$$

where  $g_2(r) \equiv \langle \psi_1^\dagger(r) \psi_1^\dagger(0) \psi_1(0) \psi_1(r) \rangle / \langle \psi_1^\dagger(0) \psi_1(0) \rangle^2$ ,  $f_{ij}(q_T)$  is the  $i$ - $j$  scattering amplitude evaluated at the system's characteristic momenta  $\hbar q_T$ , and  $\sigma_1 \equiv \langle \psi_1^\dagger(0) \psi_1(0) \rangle$  is the 2D density. Here  $q_T = \sqrt{mk_B T}/\hbar$  is proportional to the thermal momentum, where  $k_B$  is Boltzmann's constant. In a dilute thermal gas  $g_2(0) = 2$ . A similar formula holds in 3D, where  $f_{ij} = 4\pi a_{ij}$  with  $a_{ij}$  the 3D scattering length.

In quasi-2D, where kinematics are 2D but the 3D scattering length  $a$  is much less than the perpendicular confinement length, we can construct a 2D interaction that reproduces the low-energy scattering properties. Assuming harmonic confinement with oscillation frequency  $\omega_{\text{osc}}$ , the effective 2D scattering amplitude is [12]

$$f_\eta(q_T) = 2\sqrt{2\pi} \frac{\hbar^2}{l/a_\eta - (1/\sqrt{2\pi}) \log(\pi q_T^2 l^2)}, \quad (3)$$

where  $l = \sqrt{\hbar/(m\omega_{\text{osc}})}$  is the length scale of the  $z$ -axis confinement of the hydrogen gas. Following Ref. [1]'s discussion, the characteristic length scale for the confinement in their experiments is  $l_0 = \sqrt{2\pi}l = \hbar/\sqrt{2mE_a} \sim 5 \text{ \AA}$ , where  $E_a$  is the adsorption energy of the hydrogen on the helium film. If  $\{a_{11}, a_{12}\} \ll l_0$  Eq. (2) with the scattering amplitude from Eq. (3) reduces to the simpler form

$$\delta\omega = \frac{4\pi\hbar^2}{m} g_2(0) (a_{12} - a_{11}) \frac{\sigma_1}{l_0}, \quad (4)$$

as for a 3D gas with density  $\sigma_1/l_0$ .

The spectral shift predicted by Eq. (4) is 100 times larger than experiment. We will show that this discrepancy is consistent with the corrections found by including the

hydrogen-hydrogen interaction mediated by the helium film,  $V_{\text{med}}$ . Although  $V_{\text{med}}$  is state independent, it alters  $\delta\omega$  by reducing the probability that two particles are close enough to feel the spin dependent  $V_{ij}$ . Remarkably, even an attractive  $V_{\text{med}}$  can reduce this overlap [13].

*Helium film-mediated interaction.*—Wilson and Kumar derive a hydrogen-only effective action [14] assuming that (i) the helium film's excitations are noninteracting, (ii) the hydrogen-helium interaction potential is modeled as a van der Waals potential,  $V_{\text{H-He}}(r) = -(6\Lambda_0/n\pi)r^{-6}$  with  $\Lambda_0$  controlling the potential's strength,  $n$  is the helium density, and  $r$  is the 3D distance between the hydrogen and helium atoms, (iii) the hydrogen confinement length  $l_0$  is significantly smaller than the hydrogen-helium separation  $\zeta$ , and (iv) retardation effects can be neglected. We define the total hydrogen density operator  $\rho_t(\mathbf{r}) \equiv \sum_j \psi_j^\dagger(\mathbf{r}) \psi_j(\mathbf{r})$ , where  $\mathbf{r}$  is the atomic coordinate projected onto the plane of the 2D film. Then Fourier transforming Ref. [14]'s effective Hamiltonian gives

$$H_{\text{eff}} = H - \frac{1}{2} \iint d^2\rho d^2\rho' \rho_t(\mathbf{r}) \rho_t(\mathbf{r}') V_{\text{med}}(\mathbf{r} - \mathbf{r}') \quad (5)$$

with mediated pair interaction

$$V_{\text{med}}(\mathbf{R}) = V_0 \bar{V}_{\lambda/\xi}(\mathbf{R}/\lambda), \quad (6)$$

$$V_0 \equiv \frac{2\delta^2}{\pi^3 \lambda^2 M C_3^2}, \quad (7)$$

defining  $\delta \equiv 6\Lambda_0 \phi_g / n \zeta^4$ ,  $\lambda \equiv \sqrt{\frac{1}{M C_3^2} (\frac{1}{2M} + \frac{\beta d_0}{n})}$ , and  $\phi_g \equiv \sqrt{n d_0}$ , where  $C_3$  is the film's third sound speed,  $d_0$  is the helium film thickness,  $M$  is the helium mass, and  $\beta$  is the film surface tension (estimates of parameters in experiments are given later). The nondimensionalized potential  $\bar{V}_\xi$  is found to be

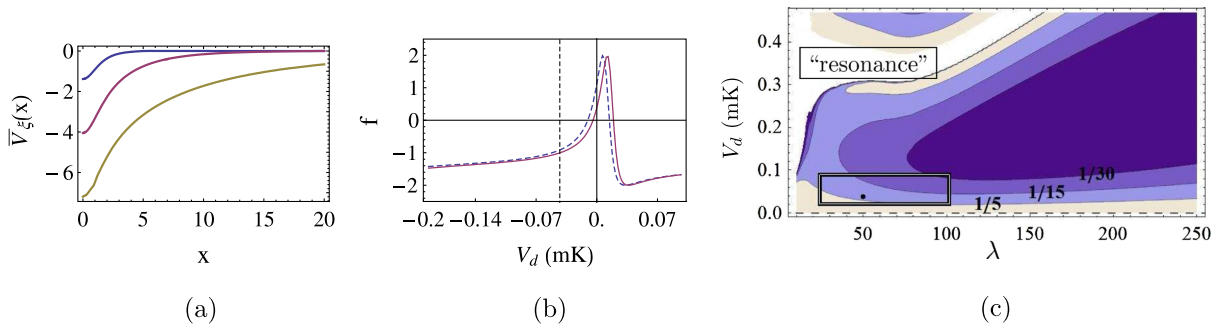


FIG. 1 (color online). (a) Rescaled mediated potential as a function of interparticle separation divided by  $\lambda$ , as given by Eq. (8), with  $\xi = 1, 4, 15$  from top to bottom. (b) Scattering amplitudes  $f$  as a function of mediated potential depth factor  $V_d \equiv (2\delta^2)/(\pi^3 \lambda^2 M C_3^2) = V_0(\lambda/\lambda_0)^2$  with  $\lambda_0 \equiv 50 \text{ \AA}$ , for the triplet (dashed) and singlet (solid) scattering channels. Vertical lines indicate  $V_d$  for the typical parameters given in the text (dashed line) and for  $V_d = 0$  (solid). The divergences near  $V_d = 0$  have been rounded off for display. (c) Contour plot of the factor by which  $V_{\text{med}}$  reduces the frequency shift as a function of  $\lambda$  and  $V_d$ . Typical values of parameters are shown by the black dot, while factor of 2 variations comprise the interior of the black rectangle. The contour plot is hidden in the white “resonance” region (far away from the relevant regime), where the scattering amplitude diverges.

$$\bar{V}_\xi(x) = \iint d^2\bar{\rho} d^2\bar{\rho}' \bar{A}(\bar{\rho}) \bar{A}(\bar{\rho}') \bar{G}_F\left(\frac{\mathbf{x} + \bar{\rho} + \bar{\rho}'}{\xi}\right), \quad (8)$$

where  $\xi$  is an argument of  $V_\xi$ , and we define

$$\bar{A}(x) \equiv \frac{1}{(1+x^2)^3} \quad \text{and} \quad \bar{G}_F(x) \equiv K_0(x), \quad (9)$$

with  $K_0$  the zeroth modified Bessel function of the second kind. Note that  $A(\rho) = -(12\phi_g\Lambda_0/\pi n\zeta^6)\bar{A}(\rho/\zeta)$  is the hydrogen-helium van der Waals interaction at a separation  $\rho$  and  $G_F(\rho) = (n/2d_0\pi\beta)\bar{G}_F(\rho/\lambda)$  is the helium film's Green's function. We numerically compute  $\bar{V}_\xi(x)$  as a function of  $x$  and  $\xi$ . Typical results are shown in Fig. 1(a).

*Spectral line shifts with mediated potential.*—To evaluate spectral shifts via Eq. (2), we calculate the scattering amplitude of  $V_{\text{tot}} = V_{ij} + V_{\text{med}}$ . Since the range of the bare hydrogen-hydrogen potential  $r_{\text{eff}}$  is much smaller than the range of  $V_{\text{med}}$ , we may replace  $V_{\text{tot}}$  with  $V'_{\text{tot}} = V'_{ij} + V_{\text{med}}$ , where  $V'_{ij}$  is an arbitrary short range potential reproducing  $V_{ij}$ 's scattering amplitude. This replacement requires that the relative momentum  $k_0 \sim \sqrt{2mV_{\text{med}}(0)}/\hbar$  of the particles when they reach the bottom of the attractive potential  $V_{\text{med}}$  satisfies  $k_0 r_{\text{eff}} \ll 1$ , which is well-satisfied. The same conditions allow us to use a boundary condition in place of  $V'_{ij}$ , chosen to reproduce  $V'_{ij}$ 's scattering amplitude [13]. We numerically solve the resulting two-particle Schrödinger equation with potential  $V'_{\text{tot}}$ .

We use the following estimates in our calculations, taken from Refs. [1,14,15]:  $l_0 \sim 5 \text{ \AA}$ ,  $a_t = 0.72 \text{ \AA}$ ,  $a_s = 0.17 \text{ \AA}$ ,  $\lambda \sim 50 \text{ \AA}$ ,  $C_3 \sim 1 \text{ m/s}$ ,  $q_T \sim (30 \text{ \AA})^{-1}$ ,  $\delta \sim 0.265\sqrt{\hbar^2 MC_3^2/2m}$ , and  $\zeta \sim 5 \text{ \AA}$ . These are typically known within a factor of 2.

Figure 1(b) shows the  $|1\rangle - |1\rangle$  and  $|1\rangle - |2\rangle$  scattering amplitudes as a function of the mediated interaction strength, with all other parameters fixed at their typical values. The two vertical lines correspond to zero mediated potential and typical mediated potential strengths.

Figure 1(c) shows the factor by which the mediated potential reduces the spectral shift  $\delta\omega$ , relative to  $\delta\omega$  in the absence of the mediated potential, as a function of the potential depth and the characteristic length  $\lambda$ . The black box denotes the range of scattering amplitudes consistent with known parameters. A black dot indicates our best estimate of typical experimental parameters. Figure 1(c) shows that for our best estimate of parameters, the mediated potential reduces  $\delta\omega$  by a factor of 7, while a nearly 30-fold decrease is possible within the range of experimental parameters. The reductions we quote are due solely to the mediated potential. Compared with the analysis in [1], there is an additional 40% reduction of  $\delta\omega$  simply by using the more accurate formula of Eq. (2) in place of the approximation Eq. (4). Thus the observed reduction in

the cold collision frequency shift is consistent with our predictions.

<sup>3</sup>He film.—Our theory also accounts for unexpected effects of adding <sup>3</sup>He to the film's surface. Since adding <sup>3</sup>He reduces the hydrogen surface adsorption energy, one would naively expect an increase in the confinement length, decrease in density, and decrease in spectral shift. Instead, adding <sup>3</sup>He with surface density  $\sim 10^{14} \text{ cm}^{-2}$  leads to an observed 25% increase in the shift. Our theory predicts such an increase, but with larger magnitude.

The <sup>3</sup>He driven change in the adsorption energy  $E_a$  modifies the mediated interaction by increasing the confinement length  $l$  by  $\sim 2\%$ – $10\%$  and by increasing  $\zeta$ , the distance of the gas to the helium surface. The fractional changes in  $\zeta$  and  $l$  should be comparable, and we take these to be equal. We find a 200%–400% increase of frequency shifts upon adding <sup>3</sup>He.

This reduction can be understood by the following simple argument, which focuses on the dependence of  $V_0$  on  $l$ . Equation (7) shows that the mediated potential roughly scales as  $V_{\text{med}} \propto 1/\zeta^8 \propto 1/l^8$ , so a 2%–10% increase of  $l$  yields a  $\sim 15\%$ – $53\%$  decrease of the mediated potential strength, moving the dashed line in Fig. 1(b) to the right by 15%–53%, leading to a drastic increase in the shift. This simple argument omits the  $1/l$  factor in  $\delta\omega$ , due to the decreased 3D density, and  $\bar{V}_{\lambda/\xi}$ 's dependence on  $l$ , but as our more detailed calculations show these effects are insufficient to compensate.

*Validity of approximations.*—We have made a number of approximations. Here we enumerate the most important ones, and discuss which ones need to be addressed in the future through a more sophisticated theory.

The most severe approximation we made is to follow Ref. [14], and neglect retardation in the induced potential. Such effects are relevant when the phase speed of the hydrogen excitations  $\omega/k$  becomes large compared to  $C_3$  at characteristic speeds  $\sqrt{k_B T/m}$  and energies  $k_B T$  [16]. For  $T \sim 50 \text{ mK}$  as in the experiments,  $\omega/k \sim 5 \text{ m/s}$ . In comparison, typical third sound speeds are  $C_3 \sim 1 \text{ m/s}$ , so we expect retardation corrections may be significant. Including the frequency dependence in calculating the spectral shift is challenging, requiring solution of a coupled set of 2D partial differential equations for each  $\omega$ . We expect that a sharp spectral peak survives, but with reduced spectral weight.

Additionally, given the long length scale of the mediated potential, we should also critically examine the assumption that only the long-wavelength limit of the  $s$ -wave phase shift is needed to evaluate the cold collision frequency shift. Equation (2) requires that the areal interparticle distance ( $n_{2D}^{-1/2}$ ) is larger than the effective range of the potential  $R_e$ . Since  $R_e \sim 250 \text{ \AA}$  and  $n_{2D}^{-1/2} \sim 100\text{--}300 \text{ \AA}$ , the approximation with only the  $k = q_T$   $s$ -wave scattering shift may not be sufficiently accurate. Similarly, the thermal wavelength is  $\lambda_T \sim 30 \text{ \AA}$  while the cold collision

regime strictly requires  $\lambda_T \gg R_e$ . This can smear out spectral lines somewhat, although since  $\lambda_T \sim R_e$  and the potential is rather shallow, the spectral lines may remain quite sharp. Here retardation helps us, as the slower moving atoms are the dominant contributors to the spectral peak, and these have a substantially larger de Broglie wavelength—a factor of 5 for the fastest contributing atoms [17].

Finally, we have neglected mass renormalization coming from virtual desorption-adsorption processes. This approximation is well-justified since the desorption process is relatively slow [18,19], implying that the mass is renormalized by at most a few percent [15].

**Conclusions.**—In summary, we have shown that incorporating the helium surface into the theory of the hydrogen gas provides a significant renormalization of spectral shifts, in addition to a  $\sim 40\%$  reduction from previous estimates by properly treating the quasi-2D nature of the hydrogen. For a reasonable parameter range these shifts are consistent with experimental observations. In addition, we have shown that adding  $^3\text{He}$  to the helium film increases spectral shifts, in agreement with experiment and in contrast to the naive theory.

While the mediated interaction provides a consistent explanation of the experimental observations, it is likely that it is not the whole story. A 3D control experiment finds an ill-understood reduction of  $\delta\omega$  relative to theory in the 3D gas [20]. The same physics is likely to be also playing a role in 2D. Given the known uncertainty in the experimental parameters, it would be quite reasonable for the mediated interaction to be responsible for only a fraction of the observed shift.

We have found that the helium surface induces an *attractive* interaction between the hydrogen atoms. It is useful to speculate on what other physical effects this interaction can cause. For example, can it drive a mechanical instability? While thermal pressure will stabilize the gas at the temperatures currently being studied, we believe that as the temperature is lowered that a “collapse” might occur, similar to the ones seen in atomic gases [21]. Importantly, the Kosterlitz-Thouless transition requires repulsive interactions, so the mediated interaction may eliminate the possibility of achieving superfluidity without significantly altering experimental parameters, for example, by increasing the helium film thickness.

Finally, we point out possible ramifications of our theory to ultracold atomic gases and elsewhere. The key to our findings is that in quasi-2D, infinitesimal attractive interactions generate zero energy scattering resonances. These are pushed to finite positive energy by the 3D interaction, but a genuine 2D interaction easily overwhelms the effects of the 3D interaction. One can imagine our present theory applying to quasi-2D Fermi-Bose or Bose-Bose mixtures where one system mediates an interaction for the other species. Interestingly, similar physics might arise in lay-

ered systems: one layer effectively mediates an interaction in adjacent layers—incorporating these effects in a consistent manner could lead to dramatically modified interaction properties for each layer.

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In Ref. [1] we incorrectly labeled the initial atomic hyperfine state used in the experiments of Ahokas, Järvinen, and Vasiliev [2]. In our notation, the correct state is  $|1\rangle = |s_e s_n\rangle = |\downarrow\downarrow\rangle$ . We also note that Safonov, Safonova, and Lukashevich [3] theoretically argued that the contact interaction shifts should vanish based on symmetries which were not taken into account in [1,2], and that after the publication of our Letter, Ahokas, Järvinen, Shlyapnikov, and Vasiliev [4] showed that the collision shifts in 3D are also anomalously small.

While these developments make it clear that film-mediated interactions are not the dominant contribution to Ref. [2]'s experimental observation, these interactions certainly do exist and our predictions stand: for example, our discussion of the film thickness dependence of the interactions, and their impact on the mechanical stability of the hydrogen clouds. As suggested by Safonov, Safonova, and Yasnikov [5], our calculation can be applied directly to studying the collision shift in the context where both of the lowest hyperfine states are initially present.

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