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Magnetic resonance line shifts in the quantum gas of atomic hydrogen

J Ahokas, O Vainio, S Novotny, J Järvinen and S Vasiliev

Wihuri Physical Laboratory, Department of Physics and Astronomy, University of Turku,
20014 Turku, Finland

E-mail: servas@utu.fi

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Abstract

We present an experimental study of the quantum gas of atomic hydrogen in a strong magnetic field (4.6 T). The gas is compressed to densities of $\sim 10^{18} \text{ cm}^{-3}$ at temperatures of 0.2–0.5 K. We observed shifts of the electron spin resonance (ESR) lines caused by the exchange interaction in atomic collisions (clock shift) and the long-range dipolar interactions. The clock shift was found to be vanishingly small in a doubly polarized gas, in contrast to a mixture of two hyperfine states. The difference is explained by properly including quantum statistical effects in the treatment of atomic collisions. From our data, we extract the difference between the triplet and singlet s-wave scattering lengths, $a_t - a_s = 60(10) \text{ pm}$, in agreement with existing theories. At densities above 10^{17} cm^{-3} , weakly resolved structures appeared in the ESR spectra, interpreted in terms of electron spin waves. We consider exchange and dipolar interactions to possibly induce the spin waves.

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Collisions between atoms in a gas lead to shifts of atomic energy levels. As they have a direct influence on transition lines, spectroscopy becomes an effective tool in studies of atomic interactions. At low temperatures the effect of collisions is to build up the mean field interaction energy, which depends on a single scattering parameter, the scattering length a . For identical particles, quantum statistics limits the allowed scattering channels due to the spin of the colliding particles and influences the shift of spectral lines. In quantum gases this shift is known as the cold collision or clock shift (CS). The name originates from the work on hydrogen masers, where this effect was the main limitation to the further improvement of the frequency stability [1]. Later, CS was also observed in atomic fountains [2, 3], in optical excitations of atomic hydrogen [4] and in microwave and radio frequency (r.f.) transitions in alkali atom gases [5, 6]. Atomic hydrogen is the simplest atomic gas and the interaction potentials of atoms in the ground electronic state are calculated with unprecedented accuracy. However, to date there still exist serious discrepancies between theory and experiment on CS in weak magnetic fields [1, 7]. The only experimental determination of the scattering length reported so far [8] provides a rough estimate rather than an accurate value.

In this work we report on the measurement of electron spin resonance (ESR) line shifts in a gas of atomic hydrogen in a strong magnetic field of 4.6 T. Under these conditions, the atomic states are well characterized by the projections of the electron and nuclear spins (figure 1(b)). At temperatures below 0.5 K, only electron spin “down” hyperfine states, which differ by the projection of the nuclear spin, are populated. We found a very small CS in a gas containing atoms of only one of these hyperfine states [9, 10], whereas a relatively large shift was observed in a gas containing a mixture of both hyperfine states ($H\downarrow$). Several explanations for these results were suggested [11–13], which consider the effects of quantum statistics and exchange interactions in atomic collisions. We shall review these possible explanations together with our present understanding in the last part of this work. The exchange interaction for hydrogen is relatively weak compared to other atoms. Therefore, the long-range magnetic dipolar interaction becomes important, causing a line shift that is sensitive to the geometry of the system. Considering these effects, our work provides a reliable determination of the difference between the singlet and triplet s-wave scattering lengths, which is in fair agreement with existing calculations [14, 15].

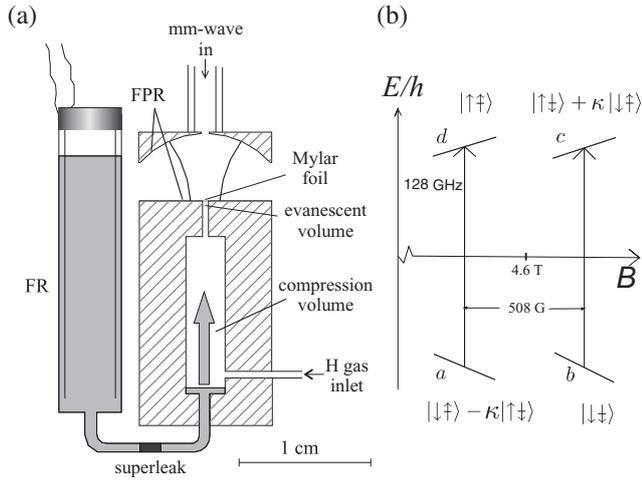


Figure 1. (a) Schematic drawing of the sample cell (FPR is the flat and spherical mirrors of the Fabry–Perot ESR resonator and FR the fountain reservoir with liquid helium level gauge). (b) Hyperfine level diagram for a hydrogen atom in a strong magnetic field with the hyperfine mixing parameter $\kappa \approx 5.5 \times 10^{-3}$.

In addition to the line shifts, exchange and dipolar interactions may lead to a propagation of spin perturbations or waves. Nuclear spin waves of spin-exchange origin were observed in quantum gases of H and ^3He [16, 17]. Dipolar or magnetostatic waves are known for ferromagnets and solid ^3He [18, 19]. In this work, we observe weakly resolved structures in the ESR spectra suggesting the appearance of electron spin waves. However, considering exchange and dipolar interactions as possible causes, neither mechanism can currently quantitatively explain the recorded spectra.

The CS in a cold gas results from the difference between the mean field interaction energy in the initial (i) and final (f) states of an atom interacting with an r.f. field. Based on the mean field theory, the clock (contact) shift for binary atomic collisions is described by [20, 21]

$$\hbar \Delta\omega = g_2(\lambda_f - \lambda_i)n, \quad (1)$$

where $\lambda_{f,i} = 4\pi\hbar^2 a_{f,i}/m$ is the coupling parameter for the i–i and f–i interactions, with $a_{f,i}$ being the s-wave scattering lengths, g_2 is the two-body local correlation for the colliding atoms and n is the gas density. If the gas contains two different hyperfine states, one has to sum over all intrastate and interstate collision contributions. However, this model does not take into account the influence of the spin of colliding atoms and thus can be applied only to optical transitions, such as the 1s–2s transition used to detect Bose–Einstein condensation (BEC) in atomic hydrogen [4]. We therefore have to consider an appropriate symmetrization with respect to the interchange of identical particles in order to extend this description to magnetic resonance transitions. For instance, in the case of fermions, scattering with zero angular momentum is forbidden by the Pauli exclusion principle so that CS vanishes [6]. On the other hand, in most cases one may disregard the internal spin structure and reduce the system to a simple two-level model, as realized for hydrogen masers and alkali atoms in zero field [5]. This simplified approach fails to explain the ESR line shifts observed in our work in a strong magnetic field and has to be appropriately modified as discussed below.

We are studying the hyperfine transitions of ground electronic state atomic hydrogen gas in a high magnetic field using ESR spectroscopy at 128 GHz [9]. At temperatures of $\lesssim 0.5$ K the strong magnetic field (4.6 T) enables us to easily realize an electron and nuclear spin polarized H gas sample, which occupies with more than 99% the single hyperfine state b ($\text{H}\downarrow\downarrow$, figure 1(b)). However, under these conditions, a precise spectroscopic measurement also becomes more sensitive to field inhomogeneities, which broaden the transition lines and hide the effects of atomic interactions, in particular the small collisional line shifts. Large gas densities are required in order to make the shifts discernible. This enhances the absorption of the r.f. power compared to other losses in the cavity and causes a distortion and broadening of the resonance lines [22]. We have thus specifically designed a sample cell (figure 1(a)), which solves these problems. In this setup the hydrogen gas is hydraulically compressed into a 0.5 mm diameter channel located in the lower mirror of the Fabry–Perot resonator and separated by a thin (12 μm) Mylar foil from the resonator volume. An evanescent r.f. field extending into the channel by the characteristic distance equal to the channel radius weakly couples the gas to the resonator. This reduces the filling factor of the sample in the cavity to $\gtrsim 10^{-3}$ with the cavity response remaining linear up to gas densities $n \approx 10^{18} \text{ cm}^{-3}$.

A detailed description of the filling and compression procedure for H gas can be found in [10] so that, here, only a summary of the steps will be given. The filling and compression of H gas in the sample cell is controlled by adjusting precisely the helium level in the compression volume with the help of a fountain valve. The helium level there is constantly monitored with a resolution of $\approx 30 \mu\text{m}$ via a capacitive level gauge in the fountain reservoir. Once the H gas inlet is closed by liquid helium, the hyperfine level population of the trapped hydrogen atoms is mainly determined by the recombination rates. Starting at roughly equal densities of a and b states ($\approx 10^{16} \text{ cm}^{-3}$, $T = 200\text{--}420$ mK), atoms in state a recombine much faster than those in the doubly polarized state b, creating a gas of high polarization $p = n_b/n_a$. The polarization ratio p depends strongly on the gas temperature and density, increasing from ≈ 10 at 300 mK to $\gtrsim 100$ at $T \approx 200$ mK. Typically, we can vary the compression factor up to the maximum value of ≈ 200 by stabilizing the helium level at different heights in the compression or evanescent volume. Once the H gas is compressed we allow the sample to decay via recombination. The decay of the sample density and of the pressure are both recorded by the decrease of the helium level in the fountain reservoir as well as by the reduction of the ESR line integrals. Measuring the change of the hydrostatic head during the decays thus enabled us to perform an absolute density calibration of the ESR absorption integrals. Within about 10% it coincides with the calorimetric calibration where the recombination heat is integrated during the decay.

Below densities of 10^{17} cm^{-3} the ESR line shape becomes particularly sensitive to inhomogeneities of the static magnetic field, limiting the narrowest possible line width to ≈ 70 mG, independent of the gas density. Increasing the gas density we not only observed a broadening of the ESR line, but also a modulation of the line shape. Several narrow

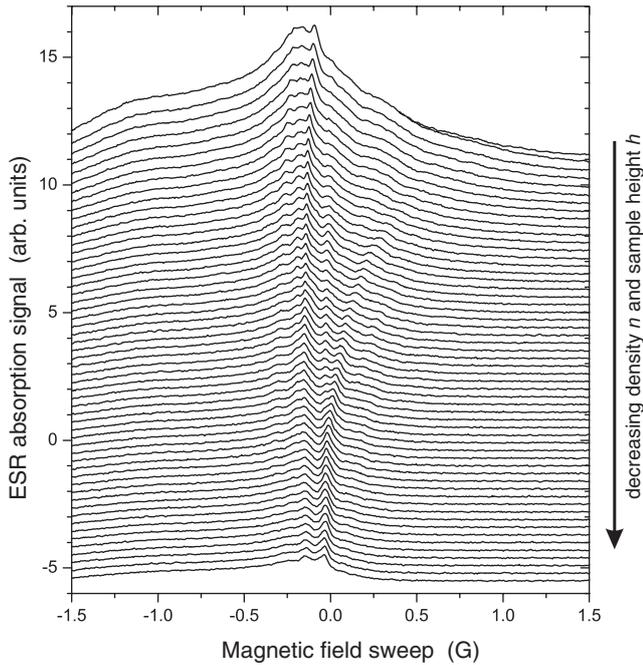


Figure 2. Electron spin waves in a magnetic field gradient of 80 Gm^{-1} . The spectra evolve from the top to the bottom, with the highest density n and sample height h at the top.

structures, varying in signal strength, appeared at positions that changed with density n and sample height h (figure 2), limiting the accuracy of the line position measurement to $\approx 20 \text{ mG}$ at densities $n \approx 10^{18} \text{ cm}^{-3}$. We suggest that these oscillations originate from electron spin waves. A similar behaviour of the nuclear magnetic resonance lines has been observed previously in $\text{H}\downarrow$ gas and was explained in terms of nuclear spin waves [16]. The origin of this phenomenon is due to the identical spin rotation (ISR) effect, which occurs when the atoms approach each other on the distance of the order of the thermal de-Broglie wavelength [23]. The frequency shift of the ISR spin wave peaks can be found from the dispersion relation [24] $\Delta\omega \sim (k_l v_T)^2 / \Omega_{\text{int}}$ with $\Omega_{\text{int}} = 4\pi\hbar a_l n / m$ being the exchange interaction frequency, v_T the thermal velocity and $k_l = 2\pi l / h$ the wave vector of the spin wave mode ($l = 1, 2, \dots$) confined in a cylinder of height h . Taking the typical values of $h \sim 0.1 \text{ cm}$ and $n = 10^{17} \text{ cm}^{-3}$ used in our experiments, we estimate the possible characteristic mode splitting in the ESR spectrum $\Delta B = \Delta\omega / \gamma_e \sim 4 \text{ mG}$.

As a second explanation we considered magnetostatic or Walker modes first found for ferromagnets [18] and later studied in solid ^3He [19]. In this case the long-range dipolar interaction leads to collective oscillations, which depend in a complicated way on the shape of the sample and excitation strength. The dipolar origin of the spin waves seems to be reasonable for the case of the ESR because of the larger dipolar moment of the electron compared with that for the proton. However, the shift of the corresponding peaks in the ESR spectrum cannot exceed the static magnetization of the sample $4\pi|\mathbf{M}| = 4\pi\mu_B n \lesssim 0.1 \text{ G}$ at the maximum densities reached in this work. The splitting between the peaks seen in figure 2 exceeds 0.5 G , in contradiction to the ISR or magnetostatic origin of the spin waves considered above.

Further studies of the electron spin waves in high-density $\text{H}\downarrow$ gas are needed to understand the nature of this phenomenon.

The measurements focused on the shifts of the $a \rightarrow d$ and $b \rightarrow c$ hyperfine transitions in hydrogen gas samples prepared at different polarizations p . Starting at the lowermost temperatures $T \approx 200 \text{ mK}$ we first realized an almost fully doubly spin polarized sample with an a state density well below our detection limit of $5 \times 10^{15} \text{ cm}^{-3}$. Within the resolution of our experiment, we did not observe any dependence of the $b \rightarrow c$ transition frequency on the gas density, similar to the result reported in our work on 2D hydrogen gas [9].

In the next step we studied the $a \rightarrow d$ transition. To make it discernible from the noise, we increased the a state density by varying the rate and strength of the compression and the cell temperature, but still remaining in the limit of $n_b \gg n_a$. The large frequency difference between the $a \rightarrow d$ and $b \rightarrow c$ transitions does not allow us to simultaneously measure both lines, as a magnetic field sweep of 507 G is required under the present experimental conditions (figure 1(b)). This would induce strong eddy current heating and magnetic field instabilities, inhibiting stable conditions and precise measurements of the transition lines. Therefore we recorded independently the decay of each state in identical compression sequences, keeping the same starting conditions.

Compared to a purely doubly polarized sample, atoms in a gas containing a mixture of hyperfine states will experience both intrastate (b - b) and interstate (a - b) collisions. Consequently, these can also contribute to the total collisional line shift, in proportion to the densities of both states. For the $b \rightarrow c$ transition the ESR line shift is then expressed by $\Delta B_{bc} = C_{bc,b} n_b + C_{bc,a} n_a$ with the proportionality constant $C_{bc,b}$ ($C_{bc,a}$) describing the influence of b - b (b - a) atom collisions on the $b \rightarrow c$ line position. From our first measurement with doubly spin polarized gas, we found the intrastate contribution $C_{bc,b}$ to be small. In order to determine the influence of the a state atom collisions on ΔB_{bc} , we succeeded in temporarily increasing the a state density by enhancing the $b \rightarrow a$ relaxation process in a rapid compression of the sample. During the initial part of the subsequent density decay, n_a decreases by an order of magnitude, whereas n_b decreases only by $\approx 20\%$ (see figure 2 in [10]), amplifying the influence of the a state atoms on the observed line shift ΔB_{bc} . The measured $b \rightarrow c$ frequency shift is presented in figure 3 as a function of density. We clearly see that the line shift induced by a state atom collisions ($\Delta B_{bc}(n_a)$) is much larger than the one due to b states ($\Delta B_{bc}(n_b)$). The corresponding proportionality constants are extracted from linear fits, obtaining $C_{bc,a} = 8(2) \times 10^{-19} \text{ G cm}^3$ and $C_{bc,b} = 5(5) \times 10^{-20} \text{ G cm}^3$, respectively.

Similarly, the $a \rightarrow d$ line shift is expressed by $\Delta B_{ad} = C_{ad,b} n_b + C_{ad,a} n_a$, with the last term being negligibly small for $n_b \gg n_a$. To optimize the measurement of ΔB_{ad} , we applied a slower and stronger compression of the sample, yielding a high b state density with a low but easily detectable population of a state atoms during the density decay (figure 2 in [10]). From the data plotted in figure 3 we extract $C_{ad,b} = 7(1) \times 10^{-19} \text{ G cm}^3$, which is within the error bars equal to $C_{bc,a}$ (see also figure 4). Repeating these measurements we reach a $\pm 20\%$ reproducibility for $C_{ad,b}$ (different triangles in

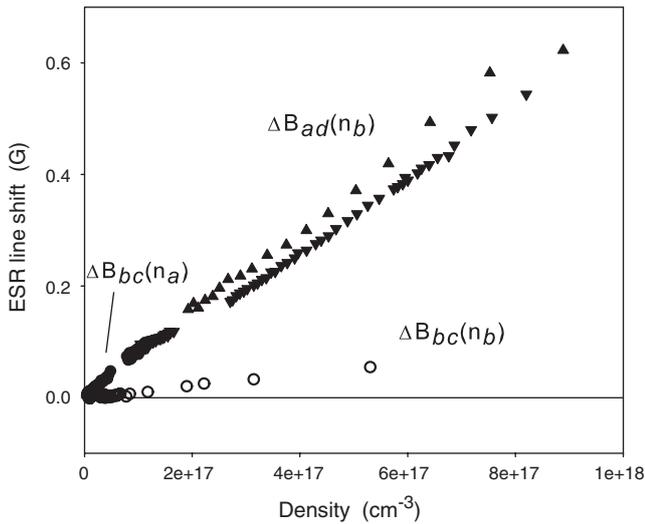


Figure 3. Measurement of the CS; ●, ▲—interstate shift $\Delta B_{bc}(n_a)$, $\Delta B_{ad}(n_b)$ (different triangles correspond to different measurements); ○—intrastate shift $\Delta B_{bc}(n_b)$.

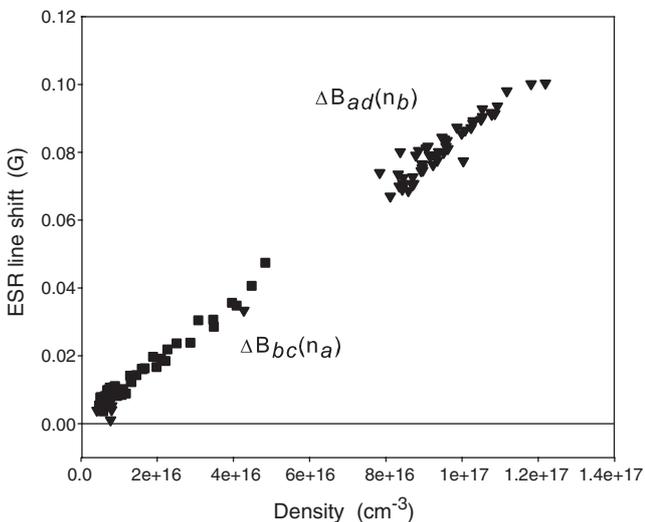


Figure 4. Measurement of the CS; ■, ▲—interstate shift $\Delta B_{bc}(n_a)$, $\Delta B_{ad}(n_b)$ (close-up of the low density range of figure 3).

figure 3). We conclude from these measurements that the CS is large in a gas with a mixture of hyperfine states, where the initially populated states differ by the projection of the nuclear spin. In contrast, we observe only a small frequency shift in a gas sample that is prepared in a single hyperfine state.

Small shifts of the ESR lines were previously also observed by us in doubly polarized (pure b-state) atomic hydrogen adsorbed on the surface of superfluid helium [9]. These results were quite surprising as numerous CS measurements in low magnetic fields with hydrogen masers and alkali vapours did not find any large disagreement with theory. We argued in [9] that the vanishing CS could be related to a reduced dimensionality. Another argument has been proposed in [25], where the reduction of the shift was explained by the interaction of hydrogen gas with ripples of the helium film. Although the above references suggest reasonable explanations for the absence of the shift in a doubly polarized gas, they do not give any hint on how to explain the relatively large interstate shift presented here.

A key point in the explanation of our results is the strong magnetic field in which the experiments were performed. In this case, the projections of electron and nuclear spins are good quantum numbers. This influences possible scattering channels for the atoms with a total electron or nuclear spin equal to 0 (singlet) or 1 (triplet). It can be shown [10, 26] that only the atoms having combinations of electron spin triplet with nuclear spin triplet, or both spins in singlet states may scatter via the channels with even angular momenta. Since at low temperatures the s-wave scattering remains the only possibility, this means that only these combinations contribute to the interaction energy. Another important issue is the symmetry of the interaction Hamiltonian between the r.f. field and the spin states of the colliding atoms, which cannot change the parity of the two-atom wave function [27]. This leads to the situation in which each atom of the pair that is exposed to the external r.f. field evolves into a superposition state between the initial and final states (e.g. b and c states for the b–c transition) so that the interatomic interaction is always via the same triplet electronic potential. Therefore, in the case of a pure b state sample there is no change of the interaction energy and in this approximation the CS vanishes. Similar arguments can be used to derive the CS in the presence of the third hyperfine state, but the result changes drastically [10]. Then the interstate shift coefficients do not vanish and can be expressed in terms of triplet and singlet scattering lengths: $C_{bc,a} = C_{ad,b} = (\pi\hbar/\gamma_e m)(a_t - a_s)$. From our data in figures 3 and 4 we can then extract the difference of the scattering lengths $\Delta a = a_t - a_s = 60(10)$ pm, which is in good agreement with theoretical calculations [15]. A larger shift by a factor of 2 has been derived in [11, 13] using basically the same arguments but treating differently the indistinguishability and correlation properties of colliding atoms. The correct determination of the difference in scattering lengths from the experimental data is part of an ongoing discussion [28, 29].

Several attempts have been made to explain the small but nonzero shift in the doubly polarized gas. For the 2D case, the authors of [13] considered the possible presence of small a state concentrations originating from two-body dipole–dipole relaxation. However, the two-body relaxation rate should vanish for the geometry in the experiments [9], when the surface is normal to the static magnetic field. Also, the surface roughness of the Mylar film, known to be very small, cannot lead to a substantial dipolar relaxation. A steady-state density of the a state should strongly depend on temperature, which has not been observed in the experiments. Another explanation suggested in [13] is related to a reduction of the adsorption energy and hyperfine constant of the adsorbed atoms at increasing gas densities. A rough evaluation of this mechanism gives a six times smaller shift than observed [13] and therefore cannot be considered as a reasonable explanation. A more plausible explanation has been proposed in [10], which concerns both the 2D and 3D cases. The authors suggested that the weak dipolar interaction may contribute not only with its long range part, i.e. due to magnetization of the gas. Scattering properties of the atoms and the coupling parameter λ may appear different for atoms with projections of the total electron spin 0 or 1. This may lead to line shifts of dipolar origin, which are dependent on

the shape of the sample. This effect is expected to be weaker for the 2D geometry used in [9], as indeed observed in the experiments.

Further experiments are designed to study more accurately the ESR in dense hydrogen gas. We hope that they will help us to clarify the nature of the line shifts and origin of the spin waves in this quantum system.

Acknowledgments

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