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Collision and Motional Averaging Effects in Cryogenic Atomic Hydrogen Masers

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Summary

Substantial progress has been made toward optimizing the performance of the neon surface cryogenic hydrogen maser, preparatory to measuring the hyperfine induced frequency shifts (HIFS) in collisions between hydrogen atoms at low temperatures. Self-excited maser oscillation has been achieved at temperatures from 8.5 to 11.5 K. There is little surface relaxation at the higher temperatures. There is substantial collision broadening at the highest achievable atom densities, which is useful for spin exchange cavity tuning and which indicates the presence of measurable HIFS. We have also made significant progress in understanding the Doppler effect in motional averaging systems such as the cryogenic maser.

Status of the Research

At the end of the last report period, we had substantially completed construction of the maser, as well as tests of the refrigeration and vacuum systems. Briefly, it operates as follows. Hydrogen molecules are dissociated in a 1 cm diameter, liquid nitrogen cooled pyrex tube by a 180 MHz rf discharge shown at the top of the figure. Atoms emerge from a 2 mm orifice in the discharge tube, and those in the upper two ground hyperfine states are focused by a hexapole state-selecting magnet toward the 2 mm orifice of the sapphire microwave cavity below. Waste atoms and molecules are cryopumped by charcoal-coated copper panels surrounding the source and magnet and cooled to 10 K by cold helium gas from a liquid helium storage dewar. In the microwave cavity, atoms are stimulated by the radiation emitted by other atoms to make transitions to the lowest hyperfine state, adding the energy of their own transitions to maintain the maser oscillation. Exhaust atoms strike a 35 K copper surface after leaving the cavity and recombine there. The oscillation power is sampled by a small loop at the bottom of the cavity. A second small cavity loop admits additional microwave signals for diagnostics, and a third loop couples in a varactor for fine tuning the cavity frequency. The cavity is surrounded by low temperature magnetic shielding and is enclosed in a vacuum can, which is immersed in liquid nitrogen. Cooling of the cavity to 7 K is accomplished by a thermal link to a cold plate cooled by cold helium gas from a second storage dewar. The temperature of the cavity is controlled by feeding back the signal from a germanium thermometer to a heater around the cavity.

Several modifications were necessary before looking for signals, the first of which was to seal the bottom of the cavity can (see Fig. 1) so that neon gas



Figure 1. Cryogenic Hydrogen Maser.

used to coat the inner surface of the maser cavity could not escape into the vacuum can and cause a conduction heat leak to the surrounding liquid nitrogen bath. Nominally non-magnetic vacuum feedthroughs from several vendors were tested and found to have significant magnetism, requiring the placement of the feedthroughs on small copper tubes protruding through holes in the magnetic shielding.

Once the cavity was sealed, we investigated techniques for coating the inner maser cavity surface with solid neon. The first step was the development of diagnostics to monitor the deposition process. Solid neon has a dielectric constant of about 1.3, compared to 9.4 for sapphire. Treating the thin layer of neon as a first order perturbation on the cavity's size, we find that 1 µm of neon condensed uniformly on the inside of the cavity decreases the TE011 resonant frequency by 510 Hz. We use a modulated microwave signal and lock-in techniques to monitor the center frequency of the cavity resonance with precision of about 3 Hz, equivalent to about 20 atomic layers of solid neon frozen on the curved inner surface of the cavity. This allows us to monitor the average neon thickness rather precisely as we form a surface. Unfortunately, the resonant frequency of the cavity indicates only the average thickness of neon on the curved inner surface of the cylindrical cavity, and provides no information about thin spots. We rely on our careful thermal design (which results in temperature uniformity of better than 1 mK on the inner surface of the maser cavity at 10K) and our surface deposition procedure (described below) to provide a uniform coating. The uniformity of the coating, which affects the wall shifts and can affect the linewidths, can be checked only after a surface has been made and is in use.

Previous experiments with solid neon surfaces¹ had found that atomically smooth 10 μ thick neon surfaces could be made by filling a hollow chamber with neon near the triple point (24.56 K and 325 T), then condensing and cooling the neon over a five hour period. Condensation at temperatures and pressures below the regime of vapor-liquid coexistence or fast cooling from the triple point, on the other hand, produced cracked, fractured surfaces. Our previous experience with frozen hydrogen and neon surfaces indicated that uneven surfaces would introduce time-dependent surface relaxation and frequency shifts. Consequently, we developed a technique for condensing liquid neon at temperatures and pressures just above the triple point, followed by slow cooling. However, in our open (state-selected beam) geometry it turns out to be crucially important to avoid flowing neon vapor from the relatively warm magnet can into the cavity orifice when the cavity temperature is below 20 K. Otherwise, solid neon is likely to build up on the first cold surface encountered, partially or completely blocking the entrance to the cavity. After much experimentation, we learned to limit such flow by pumping the residual neon out of the magnet can while slowly cooling from the triple point to the operating temperatures, using the microwave

cavity surface thickness monitor to set the neon pumping rate so that there is no further accumulation of surface.

Our first attempts to operate the maser with surfaces prepared in this manner produced pulsed resonance signals, but did not produce self-excited maser oscillation. $1/T_2$ ranged from 6 sec⁻¹ at 11 K to 50 sec⁻¹ at 8.5 K, as shown below. in Fig. 2.



Figure 2. Relaxation rate of magnetization as a function of temperature. The rate is higher at lower temperatures due to increase in the time spent on the surface. Note that the fitted exponent is about twice the measured binding energy of H to neon, consistent with a relaxation model in which both the surface fraction of atoms and their surface dwell times are exponential in the binding energy.

The large temperature dependence, exponential with about twice the binding energy, indicates a surface relaxation process. The original Harvard-SAO cryogenic maser initially experienced similar difficulties when using thin, superfluid liquid helium surfaces over bare sapphire. In that case coating the bare sapphire with Teflon alleviated the problem by lifting the adsorbed atoms farther away from magnetic impurities inside the sapphire. Following the^{:--} lead, we readjusted the coarse tuning of the maser cavity to permit use of thicker neon coatings.

Our next run achieved oscillation with a 100 μ m thick neon surface, at temperatures between 8.5 K and 11 K. As hoped, the relaxation problems noted in the earlier runs were much improved with the thicker surface: total linewidths

were as small as 0.5 Hz, close to the "geometric" linewidth 0.25 Hz expected from the average rate at which atoms exit the cavity. The maser oscillation frequency drifted at rates typically about 10 μ Hz/sec due to a variety of instabilities. The chief difficulty was that the power coupled to the discharge was unstable at high dissociation power levels, causing instability of the atomic beam. Cavity mistuning coupled the beam instabilities directly into the oscillation frequency. We observed appreciable changes of linewidth with changes of beam intensity, as indicated below in Fig. 3.



Figure 3. Oscillation power (arbitrary units) plotted against radiative linewidth as mesaured by changes of oscillation frequency with cavity tuning. T = 11K.

The maximum atom density, as indicated by the change of linewidth above from its minimum to maximum, assuming approximately the predicted collision broadening cross sections, was only about 5 x 10^{11} cm⁻³, less than our design goal by more than a factor of ten. Although the HIFS predicted for these atom densities are about a part in 10^{-12} , easily an order of magnitude larger than those we measured previously in a room temperature maser², improving the maximum available beam intensity would make the measurments much easier and would improve the maser performance as a short term frequency standard.

After these first encouraging results, we made several improvements. We improved our ability to accurately measure magnetic fields in the maser by driving the magnetic field dependent transitions. The hydrogen source stability was greatly improved by changing to inductive coupling. We found and removed some mildly magnetic spacers from the maser cavity. we renovated the hydrogen gas handling system and added a palladium purifier, in order to reduce the potential for poisoning of the hydrogen discharge by impurities. We have refined our surface formation procedure by monitoring the difference between neon vapor pressure and magnet can pressure in real time. This effort was prompted by several runs in which the cavity was blocked during surface formation due to sm ill neon flows into the cavity at low temperatures. With this refinement, we can completely form a surface above 20 K, where the risk of blocking is small, and then cool down while evaporating neon from the cavity. Operationally, we do this by pumping on the magnet can so that the magnet can pressure is always slightly lower than the vapor pressure of neon in the cavity.

In recent runs we were able to spin exchange tune the cavity. However, we continued to see frequency drifts in the cryogenic maser of a few μ HZ/sec: an improvement by a factor of three over previous runs, but still troublesome.



Figure 4. Difference in frequency between the cryogenic maser and a room temperature maser over time and with changes of cavity tuning varactor setting. The right hand figure amplifies the data at the lower frequency. At both varactor settings there is an approximately linear drift at about 2 microhertz/sec.

We believe that the small drifts illustrated above are due to drifts of the applied magnetic field because of a small dependence of the current supplies on the temperature of the room. We have recently constructed new, ultra-stable current supplies to replace the old ones. As the scatter about the drifts is only of the order to be expected from a conventional room temperature maser, we are reasonably confident of being able to make frequency comparisons over a few minutes limited only by the frequency fluctuations of our conventional reference maser.

The present stability of the cryogenic maser linewidth and frequency are such that we can now turn to several diagnostics to characterize its performance and point the way to further improvements. In the near term we plan the following:

- Using pulsed magnetic resonance, study T1 and T2 at temperatures below the minimum temperature for oscillation. This information will provide a better estimate of the atomic flux and also some information about the nature of the residual relaxation on the surface.
- Study the linewidth of the oscillating maser as a function of magnetic field, in order to determine the correlation time associated with magnetic relaxation and so point to the origin of the magnetic field gradients.
- Evaluate the short term and long term frequency stability.
- Experiment with varying the input level populations using a transverse rf coil already installed in the transition section.

Doppler effect in Hydrogen Masers

Significant progress in understanding the Doppler effect in motional averaging systems like conventional and cryogenic hydrogen masers was made (at essentially zero cost to this grant). Prior to this grant we had checked our second order random perturbation theory calculations only as applied to one-dimensional classical radiators. For his senior undergraduate research thesis Andrew Kerman extended the one-dimensional simulations to quantum mechanical calculations of the spin space density matrix for atoms interacting with an electromagnetic traveling wave. Last summer five participants in our National Science Foundation Research Experiences for Undergraduate site formed a team to extend the quantum mechanical calculations to spherical and cylindrical geometries. Briefly, the calculations compare the atomic magnetization M produced by an electromagnetic traveling wave in a sample of atoms bouncing randomly around in a cell, as predicted by second order perturbation theory and as simulated using Monte Carlo techniques. The perturbation theory result turns out to be in excellent agreement with the simulations in the limit of vanishing electromagnetic field amplitude x and to deviate as x increases roughly as dM/M $\approx 2 \text{ x}^2 \text{ tc}^2$, with t_c the average time for a trip across the cell. In our cryogenic maser the maximum $xt_c \approx 10^2$, so that we have confidence in using the perturbation theory to calculate motional averaging effects in our experiments. Should we or others succeed in achieving cryogenic maser oscillation at significantly higher atomic beam intensities, a more accurate theory would be needed. A promising approach which was discovered this summer, but needs to be followed up in detail, is that treating the first two trips across the cell separately greatly increases the agreement between perturbation theory and simulations. Another senior research thesis student is continuing this work.

Publications

The basic ideas motivating this research have already been published 3-5. We will publish a description of oscillation in our new maser in Physical Review A. Our Doppler effect work will also be published in Physical Review A.

Personnel

During the period of work reported here Professor Stuart B. Crampton, Principal Investigator, participated fully. He received his Ph. D. from Harvard University in 1964, with a thesis entitled <u>Hyperfine and Spin Exchange</u> <u>Experiments with the Atomic Hydrogen Maser</u>. From August 1, 1991 through June 1992 John H. Krupczak participated full time as a graduate research assistant. He possesses M. S. and Ph. D. degrees, awarded in 1986 and 1993 by the University of Massachusetts at Amherst. His M. S. dissertation is entitled <u>An</u> <u>Experiment in Earth Seasonal Solar Energy Storage</u>, and his Ph. D. thesis <u>Design</u> <u>and Construction of a Cryogenic Hydrogen Maser</u>, based on his work in this lab. Dr. Donald McAllaster participated in this work half time October and November 1991, and full time thereafter. His Ph. D. was awarded him in 1992 by the University of Massachusetts at Amherst, and his thesis was entitled <u>Spin</u> Transport in Dilute, Spin-Polarized Solutions of Helium-Three in Helium-Four.

Interactions

Donald McAllaster presented a poster on the cryogenic maser at the annual meeting of the Division of Atomic, Molecular, and Optical Physics in May 1993 in Reno, NV. Stuart Crampton presented a poster on the same subject at the Gordon Conference on Atomic and Molecular Physics, held in Wolfeboro, NH in July 1993.

Discoveries, Inventions Patent Disclosures

None.

Evaluation of Progress and Accomplishments

We have made good progress in the second year of AFOSR support. We have demonstrated oscillation of our cryogenic maser with greatly improved temperature, surface relaxation and magnetic field stability, as compared to its prototype predecesor.⁶ We have made some progress in optimizing its performance and are appraoching stability sufficient to make the HIFS measurements. AFOSR support has been critical to the success of this research. Indeed, the work will soon cease if we are not able to replace it.

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