N O T I C E

THIS DOCUMENT HAS BEEN REPRODUCED FROM MICROFICHE. ALTHOUGH IT IS RECOGNIZED THAT CERTAIN PORTIONS ARE ILLEGIBLE, IT IS BEING RELEASED IN THE INTEREST OF MAKING AVAILABLE AS MUCH INFORMATION AS POSSIBLE

FINAL TECHNICAL REPORT

NASA Grant #NSG 5218

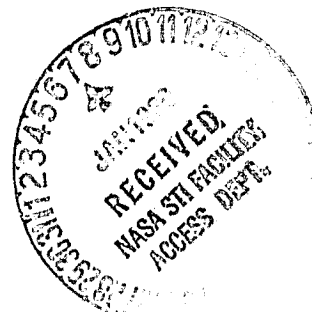EFFECTS OF HYDROGEN ATOM SPIN

EXCHANGE COLLISIONS ON ATOMIC HYDROGEN

MASER OSCILLATION FREQUENCY

December 1, 1977, through November 30, 1979

Principal Investigator:

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Introduction

Frequency shifts due to collisions between hydrogen atoms in an atomic hydrogen maser frequency standard affect the precision of hydrogen standards under some conditions. Investigations of frequency shifts proprotional to the spin exchange frequency shift cross section λ and those proportional to the duration of exchange collisions were the first subject of study under this grant. Midway through the grant the focus of the work shifted to the feasibility of operating a hydrogen frequency standard at liquid helium temperatures.

Part One

Spin Exchange Frequency Shift Cross Section

Collisions between hydrogen atoms radiating on the ground state hyperfine transitions in an atomic hydrogen maser shift the frequencies of the transitions because of two distinct mechanisms which affect the oscillation frequency of a hydrogen maser frequency standard in slightly different ways. Electron exchange during collisions transfers coherence from one radiating atom to another with a slight phase shift.[1] Continual shifts of phase produce a frequency shift proportional to a spin exchange frequency shift cross section λ and also dependent on the populations of the hydrogen atom levels involved in the frequency standard transition. The phase of oscillation of an atom radiating on the hyperfine transition is further retarded by electron exchange collisions because the exchange operator only partially commutes with the hyperfine interaction operator. To the extent that these operators do not commute, the hyperfine interaction phase falls behind during a collision by an amount proportional to the duration of the collision and only very weakly dependent on level population.[2]

The nature of the dependence of the frequency shift proprotional to λ on level populations is such that the frequency of a hydrogen maser operating as an active frequency standard is shifted by an amount proprotional to the degree to which the maser microwave cavity is mistuned less an additional term proportional to λ, the difference between the two terms determining the next shift. By carefully adjusting the microwave cavity frequency so that there is no variation of maser oscillation

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frequency as the atomic beam intensity is varied, the two shifts due to cavity mistuning and the collisional shifts proprotional to λ can be made to cancel,[3] leaving only the shift proprotional to collision duration. Careful invesitgation of the microwave cavity frequency for which these two shifts cancel provides, in turn, a measure of λ and checks that present theoretical understanding of frequency shifting mechanisms in hydrogen masers do indeed account for the behavior observed. A previous measurement of λ was in agreement with the theoretical predictions of an adiabatic treatment of spin exchange collisions, but in the course of those measurements two hitherto unsuspected frequency shifting mechanisms were discovered and eventually accounted for. [4,5] Subsequently, a more stringent test of the theoretical prediction for λ was devised and put into motion.[6] Preliminary results were in disagreement with theory.

The method consists of comparing the variation of hydrogen maser oscillation frequency with atomic beam intensity for atoms stored in either of two Teflon-coated quartz envelopes both within the same maser microwave cavity. The geometrical filling factor F which measures the average value of RF magnetic field (squared) in a particular storage region relative to the energy stored in the cavity is different for the two envelopes, and consequently, the value of cavity tuning for which cavity mistuning shifts cancel shifts proportional to λ is slightly different by an amount proportional to λ. Deriving λ from the observed change of cavity tunign requires accurate knowledge of the filling factors for the two regions, and these must be calculated numerically, given detailed understanding of the geometry and the ways in which the quartz envelopes distort the RF cavity fields. The geometry of the

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cavity and the storage envelopes had been designed to give a change of cavity tuning of about 30 Hz with a cavity width of about 40 KHz. Preliminary measurements indicated zero change of cavity tuning within precision of about 4 Hz. However, the variation of cavity tuning with ambient magnetic field suggested that atoms were leaking from the storage envelope to which they had been originally directed by the beam switching device into the other storage envelope. Calculations indicated that leakage to the extent observed could significantly affect the results. In addition, a slight shift of the cavity tuning was observed when the beam switching device was activited, and there was concern that the change might not be tracked precisely by our cavity monitor. It became clear that this technique was not likely to produce a significant improvement in the measurement of λ. In consultation with the NASA technical officer responsible for this work, we decided to move on to the study of frequency shifts proportional to the duration of hydrogen atom collisions.

Part Two
Frequency Shifts Due to H-H Collision Durations

An undergraduate thesis student, Steven A. Davidson, '80, has undertaken this experiment as a senior research project. After some delays due to problems with the vacuum system, we have achieved self-sustained oscillation without Q multiplication using the storage bottle geometry described in our 1978 proposal. Linewidths with the hole in the top of the storage bottle spin range from about 2 Hz at low H beam intensity to about 3 Hz at maximum beam intensity. The next step is to install the Teflon cone. We expect to pursue this work to its conclusion dispite the termination of NASA support. There are as yet no numbers to report on the basis of the work done during the grant period.

Part Three
Storage of H Atoms at Liquid Helium Temperatures

In consultation with the NASA Technical Officer responsible for this work, we have directed most of the time and the funds supported by this grant to the study of the ground state hyperfine resonance of H atoms stored as a gas in a quartz bottle coated with frozen molecular hydrogen at 4.2 K.[7] The apparatus is essentially the same as that developed at M.I.T. during 1977-78 and described in reference 7, to which reference will be made in this report for description of the apparatus and notation.

(i) The liquid nitrogen "source" dewar (Fig. 1, B) has been replaced by a much longer one more carefully sealed at the top, so that the storage bulb is covered by much more liquid helium and so that it is possible to reduce the temperature of the liquid helium bath by pumping on its surface. We are now able to obtain stable conditions for 3 or 4 hours at 4.2 K and up to 2 hours below 4.2 K.

(ii) Silicon diode temperature sensors (Lake Shore Crytronics) calibrated to 0.1 K have been embedded in the bottom of the microwave cavity, the top of the microwave cavity, and in a copper collar which now surrounds the tube connecting the dissociator to the storage bulb.

(iii) A microprocessor-based computer has been interfaced to the pulse electronics, temperature sensing, and data acquisition.

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(iv)   A vacuum regulator (Lake Shore Cryotronics) has been
built into the line which pumps on the helium bath, so as
to regulate the vapor pressure over the bath to a few tenths
of a torr.

We have done nine liquid helium temperature experiments, and there
is a lot of data. The most important results to date are as follows:

(i)   We have found that the time constant for the storage bulb
to come to temperature equilibrium with the surface of the
helium bath, when the vapor pressure above the bath is allowed
to rise by a few hundred torr, is of order 30 minutes.   30
minutes is much longer than the time waited before taking data
at high vapor pressures last winter at MIT, so that the
temperature-dependence of the wall shift inferred then is
much too low. Our new measurements of the temperature
dependence of the wall shift from 3.8 K to 4.5 K suggest an
adsorption energy of order 30 K. That is an interesting
result, in that it brings into agreement the adsorption energy
inferred by the (roughly determined) mean surface dwell time and
that inferred by the temperature dependence of the surface
dwell time. On the other hand, the coming into agreement of
our measurements and calculations assuming a pure, uniform frozen
molecular hydrogen surface suggests that little improvement can
be expected by more careful preparation (slow cooling and
ultrasonics) of the surfaces.

(ii)   The rates $1/T_1$ at which the level populations return to
thermal equilibrium after a pulse are very much smaller than
the rates $1/T_2$ at which the oscillating magnetization decays.

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Moreover, the best fits to the longitudinal (level population) relaxation rates have the form

$$\frac{1}{3} e^{-t/T_F} + \frac{2}{3} e^{-t/T_S} \tag{1}$$

$1/T_F \simeq 2/T_S$, and both are linear in the signal amplitude as the H atom flux is varied. The coefficients 1/3 and 2/3 are predicted by a model in which the atoms are relaxed by encountering random magnetic field gradients. The linear dependence on signal amplitude implies that the rates and the signal amplitude are both linear in H atom density. The increase of relaxation with density implies a collision process, and a sharp increase of the rates as the temperature is lowered suggests that the collisions take place on the H_2 surface.

Another interesting result is that the wall shift seems to vary linearly with signal amplitude. If the signal amplitude is indeed linear in density and if the change is due to changing the surface temperature slightly due to atom recombination, then we must conclude that the recombination rate per atom is independent of density. That conclustion implies that the atoms are not only mobile on the surface, but collisions between them are essentially two-body interactions because of the very weak binding to the surface. There seems to be some very interesting physics in the interactions of the atoms with each other while bound loosely to the surface.

On the other hand, the very large frequency shifts and large temperature dependence of frequency shifts due to adsorption of H atoms on frozen H_2 surfaces near 4.2 K make it very unlikely that these

techniques will lead to an unproved hydrogen frequency standard.

The most recent results are summarized in an abstract submitted to the APS meeting in Chicago later this month and attached to this report as an appendix.

References

1. S.B.Crampton, Ph.D. Thesis, Harvard University, 1964 (unpublished).

2. S.B.Crampton and H.T.M.Wang, Phys. Rev. A12, 1305 (1975).

3. S.B.Crampton, Pnys. Rev. 158, 57 (1967).

4. S.B.Crampton, J.A.Daivier, G.S.Read and E.R.Williams, Phys. Rev. A5, 1752 (1972).

5. S.B.Crampton, E.C.Fleri and H.T.M.Wang, Metrologia 13, 131 (1977).

6. S.B.Crampton, Research Proposal submitted to NASA, 1977 (unpublished).

7. G.H.Zimmerman III, S.B.Crampton, J.S.French, W.J.Hurlin and J.J.Krupczak, to be published in Bull. Am. Phys. Soc.

Part Four
Expenditures

A.   December 1, 1977, through November 30, 1978

    (i)   Principal Investigator Salary (partial support

    while working on the 4.2 K storage of H atoms at MIT) $1416.18

    (ii)   Technician Salary   4500.00

    (iii)   Expendable Materials and Supplies   2220.53

    (iv)   Indirect Costs   2703.00

    (v)   Fringe Benefits   <u>860.29</u>

                         $11,700.00

B.   December 1, 1978, through November 30, 1979

    (i)   Principal Investigator Salary (time spent on this

    research during the fall of 1979) $5560.00

    (ii)   Expendable Materials and Supplies   2413.01

    (iii)   Indirect Costs   2914.99

    (iv)   Fringe Benefits   <u>1112.00</u>

                         $12,000.00

Part Five

Final Inventory as of November 30, 1979

Equipment Supplied by NASA or Purchased by the Contractor and Reimbursed
or to be Reimbursed by NASA.

A. Government Furnished Property

1. Experimental hydrogen maser apparatus parts

a. Pump and manifold assembly
b. State selector magnet and source assembly
c. Magnetic shield assembly
d. Upper vacuum enclosure
e. Cavity assembly
f. 3 quartz storage bottles
g. Electronics module assembly

2. Controlled Parts

a. Frequency counter (NASA experimental)   (NASA 198072)
b. Tuning recorder (NASA experimental)   (NASA 198071)
c. Crystal oscillator (Austron 1270)   (NASA 246482)
d. Transmitter (NASA experimental)   (NASA 198073)
e. Power supply (HP 6284A, SN:H1384)   (NASA 229616)
f. Dist. Syst. Eng. Ins. (5008B, AN1164)   (NASA 187599)

3. 1 Perkin-Elmer Model 203-2000 Ion Pump and Cable

B. Equipment Purchased by the Contractor under a previous NASA
contract and transferred to NASA Grant NSG 5218

a. 4 Phillips #2628990-140 HgCdZn ultraviolet
lamps without power supplies, acquisition cost: $274.93

b. 1 Granville-Phillips Model #213 Pressure
Controller, second hand, acquisition cost: 907.24

c. Varian Model #971-0003 Ionization Gauge
Controller, second-hand, acquisition cost: 302.24

d. 1 Varian 1-1/2" bakeable valve, second-hand,
acquisition cost: 232.24

e. 1 Varian Model #971-50080J nude ionization
gauge, acquisition cost: 261.52

11.

f. Matheson #B15F-679 regulator, #68-1008 trap
    and #940F valve for a cylinder of fluorine,
    acquisition cost: $343.38

g. 1 Mill Lane Engineering aluminum jacket for
    cooling the cavity of the experimental maser,
    custom made, acquisition cost: 1700.00

h. 2 Varian Model #971-5005 glass inization
    gauges on conflat flanges, acquisition cost: 424.25

i. 1 MKS #2214S-10 capacitance manometer with
    power supply and cable, acquisition cost: 751.00

APPENDIX

Abstract Submitted
for the Chicago Meeting of the
American Physical Society
21-24 January 1980

Su gested title of session in
which paper should be placed
Atomic Structure and
Transitions

Resonance Studies of Atomic Hydrogen Adsorbed on
Frozen Molecular Hydrogen Surfaces at 3.7 K to 4.6 K.
G.H.ZIMMERMAN III†, S.B.CRAMPTON, J.S.FRENCH, W.J.HURLIN,
and J.J.KRUPCZAK, Williams College.--Previous investiga-
tions[1] of the hyperfine transition in the ground state of
atomic hydrogen have been extended to higher and lower
temperatures, in order to determine the temperature
dependence of the mean surface adsorption time. Our
measurements are consistent with a mean adsorption energy
of 28(5) K for H on H_2, assuming a thermodynamic model[2]
in which the atoms migrate freely on the surface. Studies
of longitudinal relaxation processes reveal two distinct
level population relaxation rates, differing by about a
factor of two and varying linearly with signal amplitude
as the H density is changed. The low density limit of
the slow rate fixes a lower limit of about 500 atom-
surface collisions at 4.2 K without recombination.
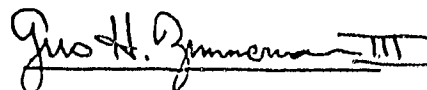
( )
(✓) Prefer Standard Session
( )

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Please place this abstract immediately
after the abstract by A. Weinrib,
Adsorption Energy of a Hydrogen Atom on a
Molecular Hydrogen Surface.
Thank you.