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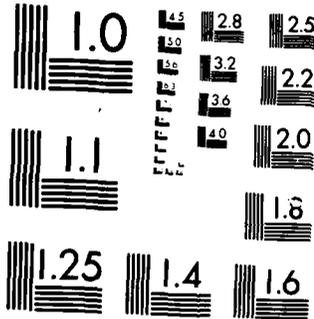
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Final Report

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ATOMIC HYDROGEN MASER INVESTIGATIONS
OF HYDROGEN ATOM INTERACTIONS FROM 4 K TO 12 K

March 1, 1980 - February 28, 1985

The President and Trustees
of Williams College
Williamstown, Massachusetts 01267

Dr. Stuart B. Crampton
Dept. of Physics
and Astronomy

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Techniques developed during previous studies of the ground state hyperfine resonance of hydrogen atoms intermittently adsorbed on molecular hydrogen surfaces were used to develop a cryogenic atomic hydrogen maser using a 5 K state-selected H atom beam and solid neon storage surfaces near 10 K temperature. The physical parameters affecting the use of such a device for frequency metrology and precision measurements of atomic parameters were determined.		

FINAL REPORT

ONR Contract # N00014-80-C-0240

ATOMIC HYDROGEN MASER INVESTIGATIONS OF HYDROGEN ATOM INTERACTIONS FROM 4 K TO 12 K

1. Principal Investigator: Dr. Stuart B. Crampton
Williams College
Williamstown, MA 01267
2. Contract Description:

The original contract called for extending downwards in temperature the techniques developed by this investigator for studying the hyperfine resonance of gaseous atomic hydrogen in storage bulbs under liquid helium. A liquid helium temperature state-selected H atom beam was to be developed and used to study the feasibility of hydrogen maser oscillation at liquid helium temperatures. As the work progressed, the contract description was modified to substitute the study of solid atomic neon surfaces at temperatures above 4.2 Kelvin, by both pulsed resonance and self-excited oscillation modes, for the study of liquid helium surfaces below 1 Kelvin.

3. Scientific Problem:

Recent experimental work motivated by prospects for observing the Bose Einstein condensation of atomic hydrogen gas at very low temperatures and high densities has produced techniques whereby useful densities of atomic hydrogen gas can be stored at low temperatures in containers coated with frozen inert gases. The relaxation times while stored are long enough for potential applications to frequency metrology and to useful measurements of atomic

collision parameters in a temperature regime particularly sensitive to the details of the interatomic potentials.

4. Scientific and Technical Approach

During the design and construction of the state-selected H atom beam we continued our studies of hydrogen atom adsorption on molecular hydrogen surfaces from 4.6 K down to 3.2 K. Using the lineshape theory that we developed, we were able to extract the adsorption energy for H atoms bound to H₂ surfaces and also the sticking coefficient (as a function of temperature over this range) of H atoms colliding with H₂ surfaces. These results are described in Annual Summary Reports SBCONR1 and SBCONR2 and in journal publications (a) and (c) listed below. The result for the adsorption energy is in good agreement with recent theoretical work. In the course of these experiments we prepared a solid atomic neon surface at 4.2 K and observed the hyperfine resonance of H atoms stored over it. We found wall shifts which were at first significantly less than for H₂ surfaces but which increased to the H₂ values with a time constant of order 1 hour as recombining H atoms built up a H₂ surface over the neon. Realizing that the lower vapor pressure of neon would allow neon surfaces to be used at higher temperatures than those possible with H₂ surfaces, that the wall shifts and relaxation rates would be exponentially less at the higher temperatures, and that H₂ might not stick appreciably to neon at the higher temperatures, we redirected the work towards studying H over solid neon surfaces at temperatures higher than 4.2 K instead of H over liquid helium surfaces at temperatures below 1 K, as planned originally. The state-selected H atom beam was designed, built, and evaluated. This work is reported in Annual Summary Reports SBCONR3 and in journal publication (b). In the final contract year the apparatus was modified so as to use the 5 Kelvin state-selected atomic beam in conjunction with neon storage surfaces maintained at temperatures up to 12 K. This work is described in Annual Summary Report SBCONR4, in journal publication (d), and in abstract (f), which is included with this report as an appendix. In a typical experiment we start with sufficient neon gas in the system as it is cooled from liquid nitrogen temperature to liquid helium temperature that the neon liquefies before freezing. Then the temperature of the neon storage surface is raised to temperatures between 8 K and 11 K, and the frequencies

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and lifetimes of H atoms radiating on the ground state hyperfine resonance are observed by pulsed magnetic resonance. During the experiment the surface is gradually lost as it evaporates. Details are given in the appendix.

5. Progress

For many of these transitory neon surfaces the wall shifts and lifetimes reproduce well as the temperature is raised and lowered between a lower limit of 8 K and an upper limit of 11 K. Below 8 K there is some evidence for buildup of H_2 over the neon, and above 11 K the neon is rapidly evaporated. The energy of binding of H to these neon surfaces reproduces well from one surface preparation to another; the effective surface area does not reproduce well, as illustrated by the variability of the intercepts in Fig. 6 of the appendix. The radiative decay rates (inverse lifetimes) and level population recovery rates are equal, indicating that the dominant relaxation process is loss of atoms, and also scatter from one surface preparation to another. Higher relaxation rates correlate with high effective surface areas. For many of these neon storage surfaces relaxation on the surface is negligible above 9.5 K compared to the rate at which atoms exit the storage bottle, and self-excited maser oscillation is easily achieved. The oscillation power level is as expected for present levels of atomic beam intensity and cavity Q. Details are given in the appendix.

This work has elucidated the basic physical parameters that determine the viability of neon storage surfaces for precision frequency metrology and fundamental measurements of collision cross sections. These are favorable. The overall size of wall shifts is an order of magnitude larger than what is to be expected for liquid helium surfaces having comparable surface-area-to-experimental-volume (A/V) ratios used at their optimum temperature of 0.5 K. However, it is easier to achieve higher A/V ratios for neon surfaces at 10 K than for liquid helium surfaces at 0.5 K, so that the choice of surface preferable for frequency metrology depends on reproducibility and stability. Unfortunately, the present apparatus is not suitable for testing the reproducibility and stability of neon surfaces. A more sophisticated apparatus in which the saturated vapor pressure of neon can be maintained over the surface as it is made and used is needed for that evaluation and for precision measurements of collision cross sections. Unfortunately, ONR support of this

work has been terminated. Work is going ahead at Harvard, funded by the Smithsonian Institution, and in Canada, funded by the U. S. National Bureau of Standards, to evaluate the reproducibility and stability of liquid helium surfaces and their suitability for precision metrology and fundamental measurements. Work on the neon surfaces, which do have potential advantages and which probe interesting physical processes at quite different and particularly interesting temperatures, awaits new funding.

To summarize the results: We have developed the first liquid helium temperature hydrogen atom beam in which the H atoms accommodate to the temperature of a molecular hydrogen surface and the saturated molecular hydrogen vapor above it. This technique is being applied already to the design of a more intense source of polarized protons for accelerator experiments. Our H beam is the first state-selected liquid helium temperature H beam. We have developed an atomic hydrogen maser operating at the lowest temperature at which self-excited oscillation has been achieved to date and at a low enough temperature to take full advantage of those improvements conjectured for hydrogen maser frequency standards at liquid helium temperatures. We have measured the parameters of hydrogen atom adsorption on 10 K neon surfaces and find them favorable for further development of a device based on these principles but equipped with better control of surface growth and stability.

6. Publications acknowledging ONR support:

A. Journals

(a) S. B. Crampton, Resonance Studies of H Atoms Adsorbed on Frozen H₂ Surfaces, J. Physics (Paris), Colloq. 41, C7-249 (1980).

(b) S. B. Crampton, J. J. Krupczak, and S. P. Souza, Progress of the State-Selected Beam Low Temperature Hydrogen Maser, J. Physics (Paris), Colloq. 42, C8-181 (1981).

(c) S. B. Crampton, J. J. Krupczak, and S. P. Souza, Temperature Dependence of Hydrogen Atom Adsorption on Molecular Hydrogen Surfaces, Phys. Rev. B25, 4383 (1982).

(d) S. B. Crampton, Hydrogen Atom Adsorption on Molecular Solid Surfaces, Ann. Phys. Fr. 10, to be published in December 1985.

B. Abstracts

(a) S. B. Crampton, G. H. Zimmerman, III, J. S. French, W. J. Hurlin, and J. J. Krupczak, Bull. Am. Phys. Soc. 25, 14 (1980).

(b) S. B. Crampton, S. P. Souza, W. J. Hurlin, and J. J. Krupczak, Bull. Am. Phys. Soc. 25, 1149 (1980).

(c) S. B. Crampton, G. H. Zimmerman, III, J. S. French, J. J. Krupczak, and W. J. Hurlin, 7th International Conference on Atomic Physics Poster Session, M.I.T., August 4-8 (1980).

(d) S. B. Crampton, K. M. Jones, G. Nunes, and S. P. Souza, Bull. Am. Phys. Soc. 29, 793 (1984).

(e) K. E. Anderson, S. B. Crampton, K. M. Jones, G. Nunes, and S. P. Souza, Proc. 17th Int. Conf. Low Temperature Phys., Part I, Elsevier Science Publishers B V. 1984, p. 539.

(f) S. B. Crampton, K. M. Jones, G. Nunes, and S. P. Souza, Proc. of the 16th Annual Precise Time and Time Interval (PTTI) Applications and Planning Meeting, United States Naval Observatory, Time Service Dept., 34th St. and Mass. Ave. N. W., Washington, DC 20390-5100, p. 339 (1984).

7. Extenuating Circumstances

The original contract was modified in order to follow up on a promising discovery. Progress has been good, and the results have justified that change of direction.

8. Unspent Funds

All ONR funds have been spent.

9. Graduate Student Degrees

There is no graduate program at Williams college. Undergraduate students associated with this work during the period of ONR support have gone on to graduate study at the University of Wisconsin, the University of Maryland, Columbia University, the University of Massachusetts at Amherst, Duke University, and Cornell University.

Appendix A

HYDROGEN MASER OSCILLATION AT 10 K

S. B. Crampton, K. M. Jones, G. Nunes, and S. P. Souza

Williams College, Williamstown, Massachusetts 01267

ABSTRACT

We have developed a low temperature atomic hydrogen maser using frozen atomic neon as the storage surface. The maser has been operated in the pulsed mode at temperatures from 6 K to 11 K and as a self-excited oscillator from 9 K to 10.5 K.

INTRODUCTION

As soon as some unpolarized hydrogen atom gas had been successfully stored at 4.2 K in a bottle coated with frozen molecular hydrogen¹, it was clear that the new low temperature hydrogen storage techniques might improve atomic hydrogen maser frequency standards.² The most likely improvement is in the short term frequency noise, which is proportional to the radiative decay rate $1/T_2$, times the square root of the thermal noise divided by the averaging time and the power radiated by the atoms.³ There is a dramatic decrease of the cross section for relaxation due to electron spin exchange collisions between the radiating atoms.⁴ Because of collisions the radiated power is quadratic in input atomic beam flux and has maximum value proportional to the inverse of the spin exchange cross section squared. The potential improvement in radiated power is large, given sufficient beam flux. Fortunately, low temperature techniques can also provide a large gain of clean, state-selected beam flux. In addition, thermal noise power in the maser cavity and receiver first stage can be greatly reduced by cooling, and substantial improvements in radiative decay rates are anticipated. These factors should combine to produce an improvement by several orders of magnitude in the short term frequency noise of hydrogen maser standards.

Low temperature techniques may also offer substantial improvements in the long term frequency stability of hydrogen maser standards. Stability to one part in 10^{15} requires temperature stability to 7.3 mK, difficult to achieve at room temperature but routinely achieved at low temperatures. Stability to one part in 10^{15} requires stability of the cavity geometry equivalent to about 1 Å in the linear dimensions; mechanical creep is literally frozen out at low temperatures. Magnetic field homogeneity and stability can be improved by using superconducting magnetic shields.

APPARATUS

Figure 1 is a schematic of the apparatus we have built to test these ideas. The apparatus is immersed in liquid helium held in a 6" ID superinsulated dewar. Molecular hydrogen is fed through a 1 cm OD pyrex tube inside a stainless steel "source dewar" separating the helium bath outside from a liquid nitrogen bath inside. The liquid nitrogen cooled 180 MHz rf discharge dissociates molecules to atoms, which pass downwards through an "accommodator" where they are cooled to 5 K and are then focused by a six pole state-selecting magnet to a 5 cm OD quartz storage bottle. The storage bottle is surrounded by a 4" OD 1420 MHz microwave cavity, a set of three 0.005" thick magnetic shields, and a vacuum tight can containing helium exchange gas, which allows the cavity and bottle to be heated uniformly to temperatures above the temperature of the liquid helium bath.

Figure 2 is a more detailed schematic of the hydrogen source and cavity assembly. Dissociated atoms pass from the discharge through a thin 2 mm ID orifice into the 5 mm ID by 1.5 cm long copper accommodator coated with solid molecular hydrogen. Semicircular baffles prevent atoms or impurities from getting through the accommodator without making about 100 collisions with its cold solid hydrogen surface. The accommodator is heated by recombining atoms or by an external heater, and it is cooled by a copper heat conduction path to the liquid helium bath. Slow moving H atoms are efficiently focused by a 1.3 cm bore by 10 cm long six pole permanent magnet through a 12 cm long by .75 cm ID entrance tube to the storage bottle. The cavity provides a uniform rf magnetic field over the storage bottle and has unloaded Q as high as 20,000 at 4.2 K.

The inside surface of the storage bottle and its entrance tube are coated with several hundred thousand layers of solid molecular hydrogen or atomic neon, frozen out from the gas phase as the apparatus is cooled initially. Care is taken to control the gas pressure and temperature so as to form some liquid first, then the solid coating. Below the threshold for oscillation a short pulse at the $\Delta F=0$ hyperfine transition frequency sets the atoms radiating on that transition, and the decaying cavity rf field is sampled by a coupling loop, converted to an audio frequency signal, and fed to the A/D converter and computer storage. The frequency and radiative decay rate are fitted directly from the signal, and level population recovery rates $1/T_1$ are determined from signals in response to multiple pulses. The signal amplitude, multiplied by T_1 , is proportional to the input flux of $F=1, m_F=0$ hydrogen atoms.

Figure 3 shows the input flux of state-selected atoms plotted against the temperature of the accommodator as measured by a carbon film thermometer. The open circles represent fluxes obtained by varying the input of atoms to the accommodator. As the flux into the accommodator increases, heat due to atoms recombining on the solid hydrogen accommodator surface warms that surface. The accommodator temperature and output flux both rise with input flux and accommodator temperature until the accommodator temperature reaches about 5.3 K, where the saturated vapor pressure of H_2 over the surface becomes high enough to impede the flow. Above 5.3 K the accommodator temperature continues to rise, but the output flux actually decreases with increasing input flux. The crosses represent fluxes obtained by leaving the input flux at the level

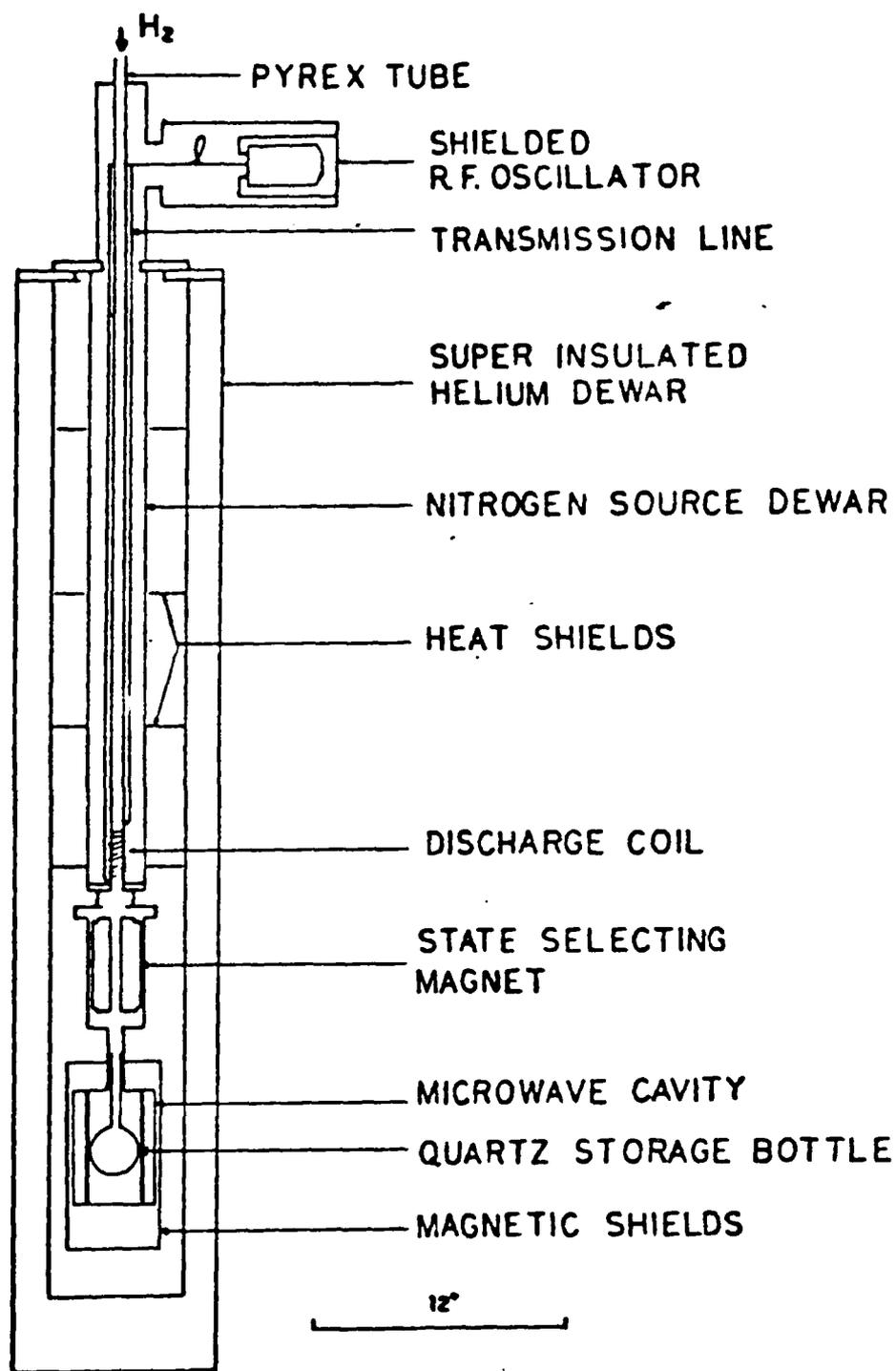


FIGURE 1 SCHEMATIC OF THE APPARATUS

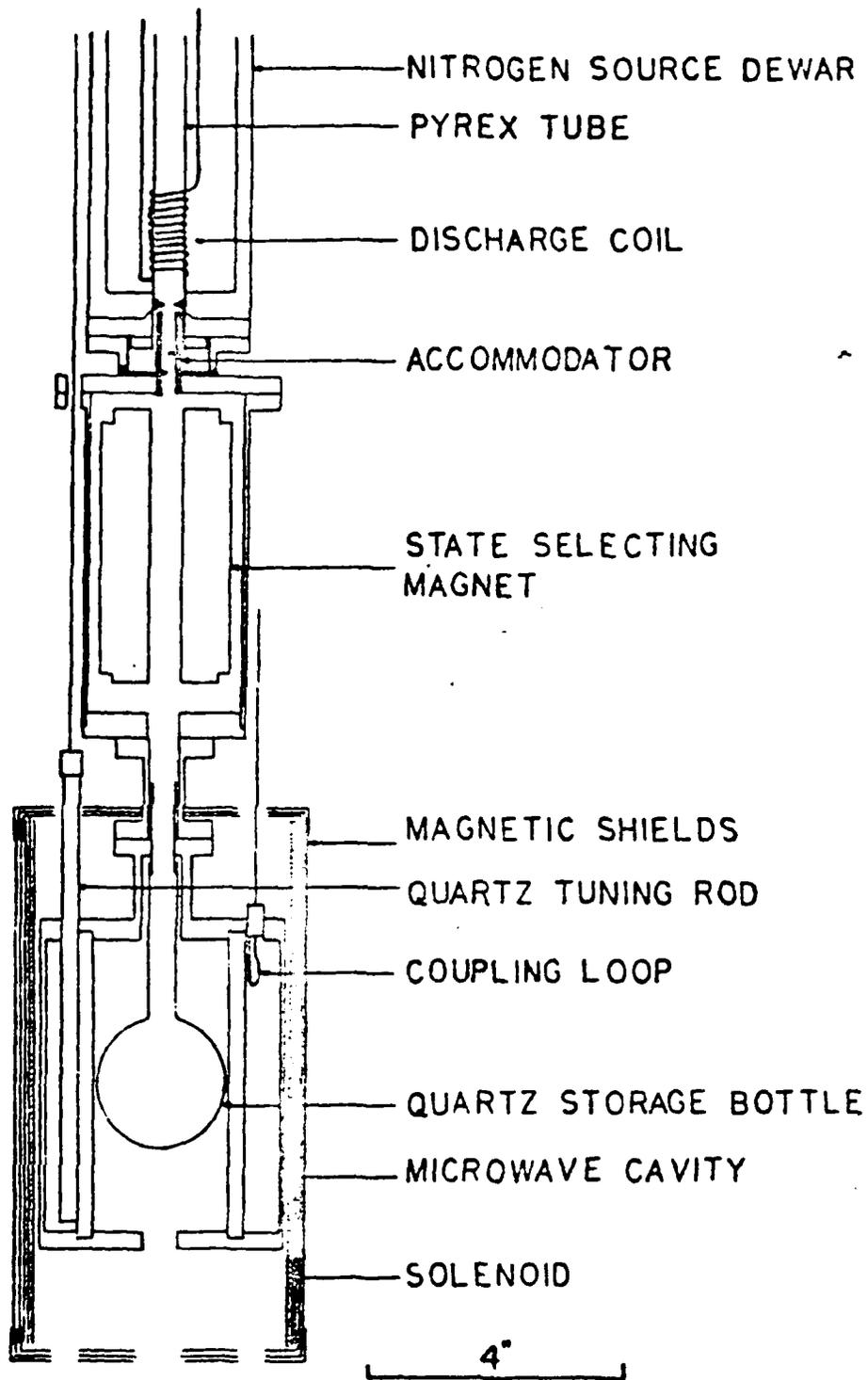


FIGURE 2 SCHEMATIC OF HYDROGEN SOURCE AND CAVITY ASSEMBLY

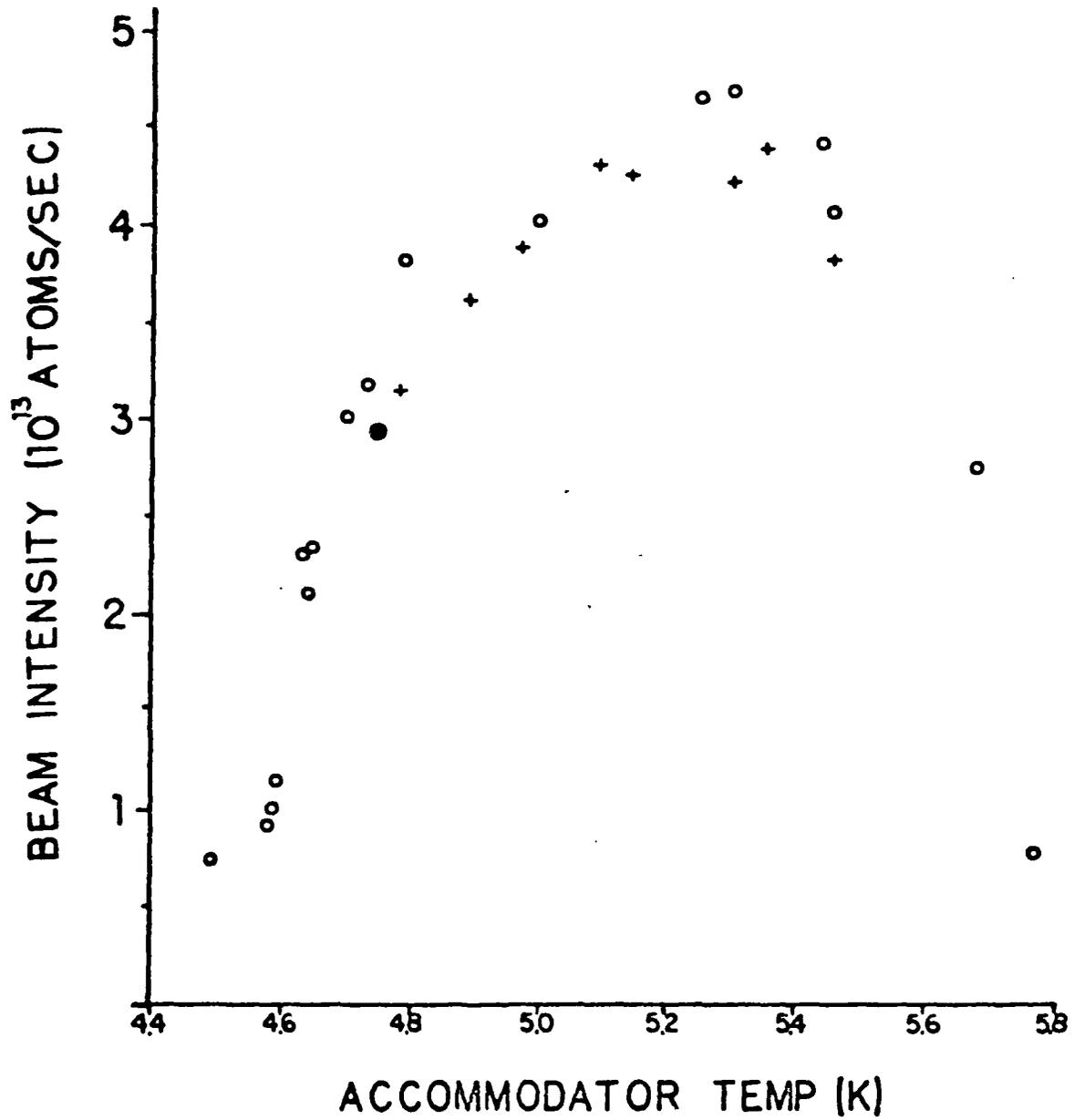


FIGURE 3 FLUX OF $F=1, m_F=0$ HYDROGEN ATOMS

that produced the flux at the solid circle and then increasing the accommodator temperature using an external heater. The coincidence of the output fluxes within errors suggests that the output flux is saturated with respect to input flux. Increasing the input flux to the accommodator will not increase output flux at the same accommodator temperature. A design requiring fewer collisions with the accommodator surface would improve output flux, but even these present fluxes compare favorably with the fluxes in room temperature hydrogen masers, and the beam is almost certainly cleaner.

SOLID MOLECULAR HYDROGEN SURFACES

We began by investigating solid molecular hydrogen wall coatings because they are the easiest coatings to make at temperatures between 4 K and 15 K and because we had already studied them over the lower temperature range from 3.5 K to 4.5 K using an earlier apparatus.⁶ Figure 4 shows some results for the wall shifts for hydrogen atoms stored over these solid hydrogen surfaces at temperatures between 4.2 K and 5.5 K. The solid line has been fit by eye with slope 35.8 K equal to the value we obtained for the binding energy of H to H₂ at the somewhat lower temperatures in the earlier work. Theoretical calculations⁷ of the binding of H to solid H₂ indicate that there should be a single bound state for motion perpendicular to the surface and almost complete freedom to move laterally. Theory⁶ predicts that for this kind of "two dimensional gas" binding the variation of wall shift with temperature should be such as to produce a straight line in Figure 4. Within errors we find that the behavior we observed at lower temperatures extrapolates well up to the highest temperatures at which we obtain signals. The data does not go higher than about 5.5 K for the same reason that the output from the accommodator drops off to very low values at accommodator temperatures above 5.5 K: the H₂ vapor over the surface of the entrance tube prevents the state-selected H beam from entering the storage bottle. At 5.5 K the lowest wall shift observed is about 50 Hz, a factor of 1000 higher than wall shifts in room temperature hydrogen masers having Teflon wall coatings. If we could go higher in temperature, the wall shifts would be dramatically lower, but the high H₂ vapor pressure prevents that.

Relaxation processes other than the dephasing while adsorbed that produces the large wall shifts can be investigated using multiple pulses to measure the level population recovery rate $1/T_1$. These rates, less the contribution from escape through the entrance, are plotted in Figure 5. A line with slope 35 K has been drawn as a guide to the behavior that would be expected for a relaxation simply proportional to the mean surface dwell time. It appears that the relaxation rates are falling off somewhat faster than at the higher temperatures. Again, if we were able to go to higher temperatures, we would expect very low relaxation rates.

To summarize the results for H₂: This pulsed resonance experiment is an interesting probe of atom-surface interactions in a particularly simple system, but it is not a candidate for precision frequency metrology because of the high H₂ vapor pressure at temperatures where the wall shifts and relaxation rates would be low.

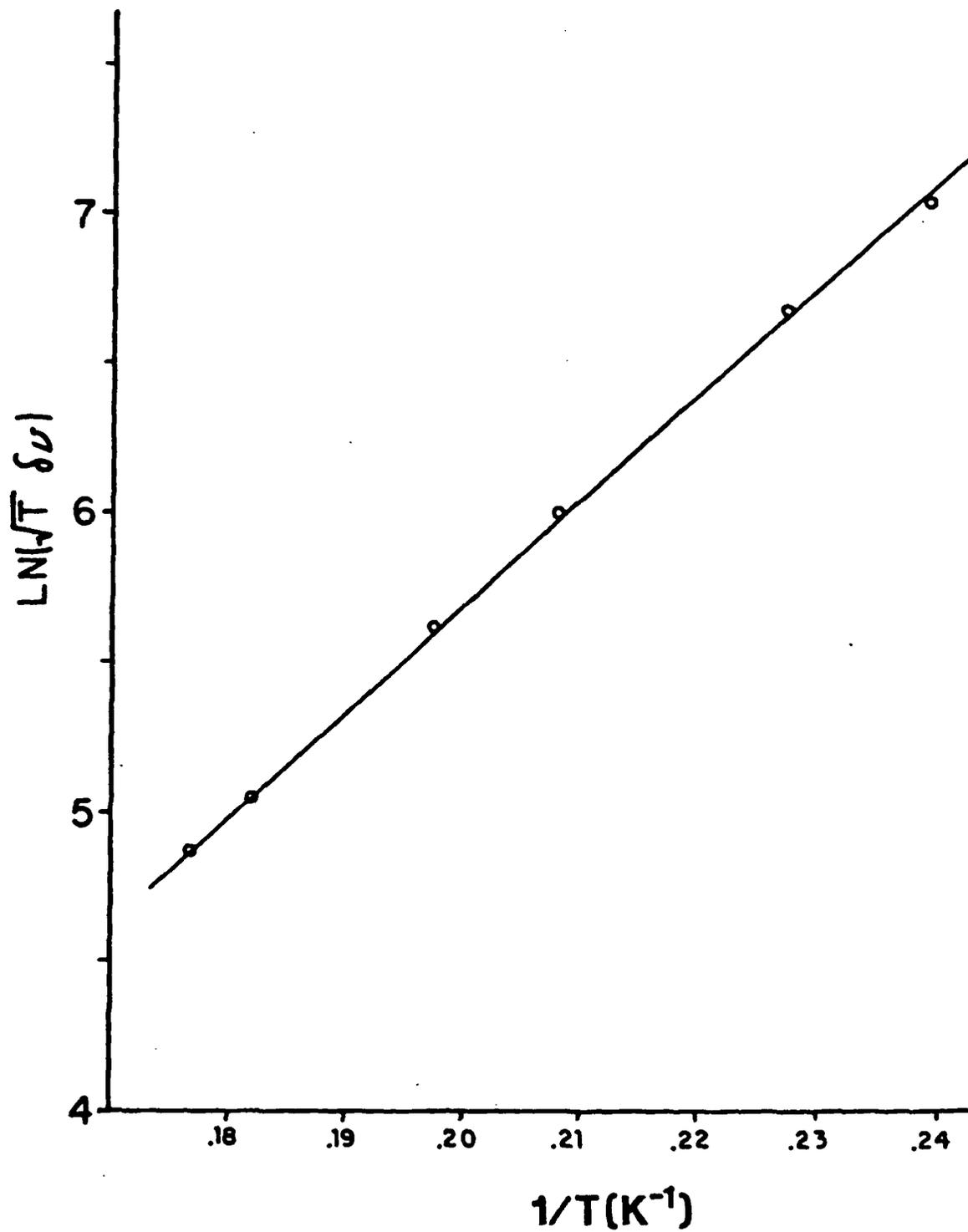


FIGURE 4 MOLECULAR HYDROGEN SURFACE WALL SHIFTS

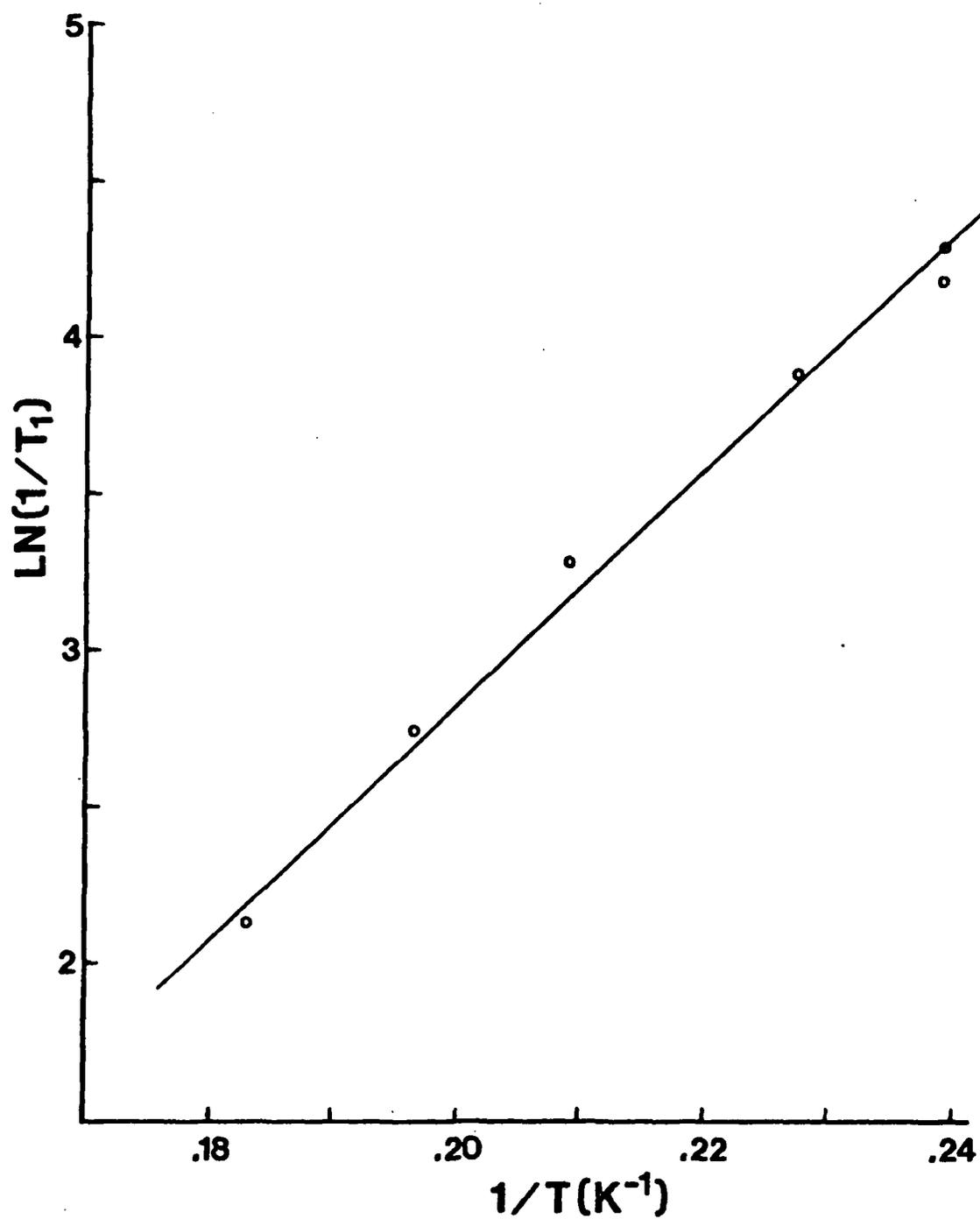


FIGURE 5 MOLECULAR HYDROGEN SURFACE RELAXATION RATES

ATOMIC NEON SURFACES

Neon atom gas has much less saturated vapor density than H_2 at the same temperature. Its electric polarizability is less than that of H_2 , so that the force between a hydrogen atom and one neon atom in the surface is less than for H_2 . Unfortunately, solid neon has less zero point motion than H_2 and so is more compact. The result is that the binding energy of H to a solid neon surface is predicted⁷ to be about the same as the binding of H to a solid molecular hydrogen surface. Figure 6 displays wall shifts we have observed for H stored in bottles with solid neon wall coatings, plotted as in Figure 4. The slopes are the same from one surface preparation to another, indicating a consistent surface binding energy in good agreement with the theoretical predictions. The intercepts vary considerably, indicating variability of effective surface area from one surface preparation to another. These data do not show indications of thin spots or contamination by some heavy impurity; both effects would shift the slope upwards as well as the intercepts. We obtain data only up to about 11 K, where the saturated vapor pressure of neon in the entrance tube prevents atoms from getting into the bottle. The wall shift at 11 K in this 5 cm diameter bottle is about 1 Hz.

The relaxation rates vary much more than the wall shifts from one surface preparation to another and even from one time to another for a single surface preparation as the surface evaporates. Evidently, the relaxation rates and the effective surface area are very sensitive to surface structure, but the binding energy is not. Within errors, $T_1 = T_2$, indicating that the relaxation process is predominantly one that removes atoms rather than relaxing them through some magnetic interaction. We have observed relaxation rates as low as 1 sec^{-1} greater than the rate of escape through the entrance, but we have also observed relaxation rates a hundred times higher. The fault lies with the high cohesive energy of solid neon, which tends to freeze into an open, snowy structure into which H atoms diffuse and eventually recombine. We believe that much more uniform and stable surfaces can be made by growing them very slowly at the temperature at which they are used and by maintaining over them the saturated vapor pressure of neon gas, but that will require a new apparatus.

MASER OSCILLATION AT 10 K

We have several times produced surfaces with low enough relaxation rates to support self-excited maser oscillation at temperatures ranging from 9 K to 10.5 K at fluxes of order 10^{13} state-selected atoms sec^{-1} and cavity Q's as low as 3500. The radiated power at this Q has been of order 10^{-12} watts. The linewidth of about 3 Hz has been dominated by the rate of escape through the bottle entrance. The short term frequency noise is less than that of our room temperature comparison maser, but we have not yet been able to measure the short term noise precisely. This first experimental apparatus is not optimized for temperature stability or magnetic field stability or even for surface stability. Although the wall shifts using solid neon wall coatings at 10 K are relatively high compared to the wall shifts in conventional hydrogen maser standards, the physics of the surface is much simpler. Whether the reproducibility and stability of the solid neon surfaces can be made to compensate for the higher wall shifts remains to be seen.

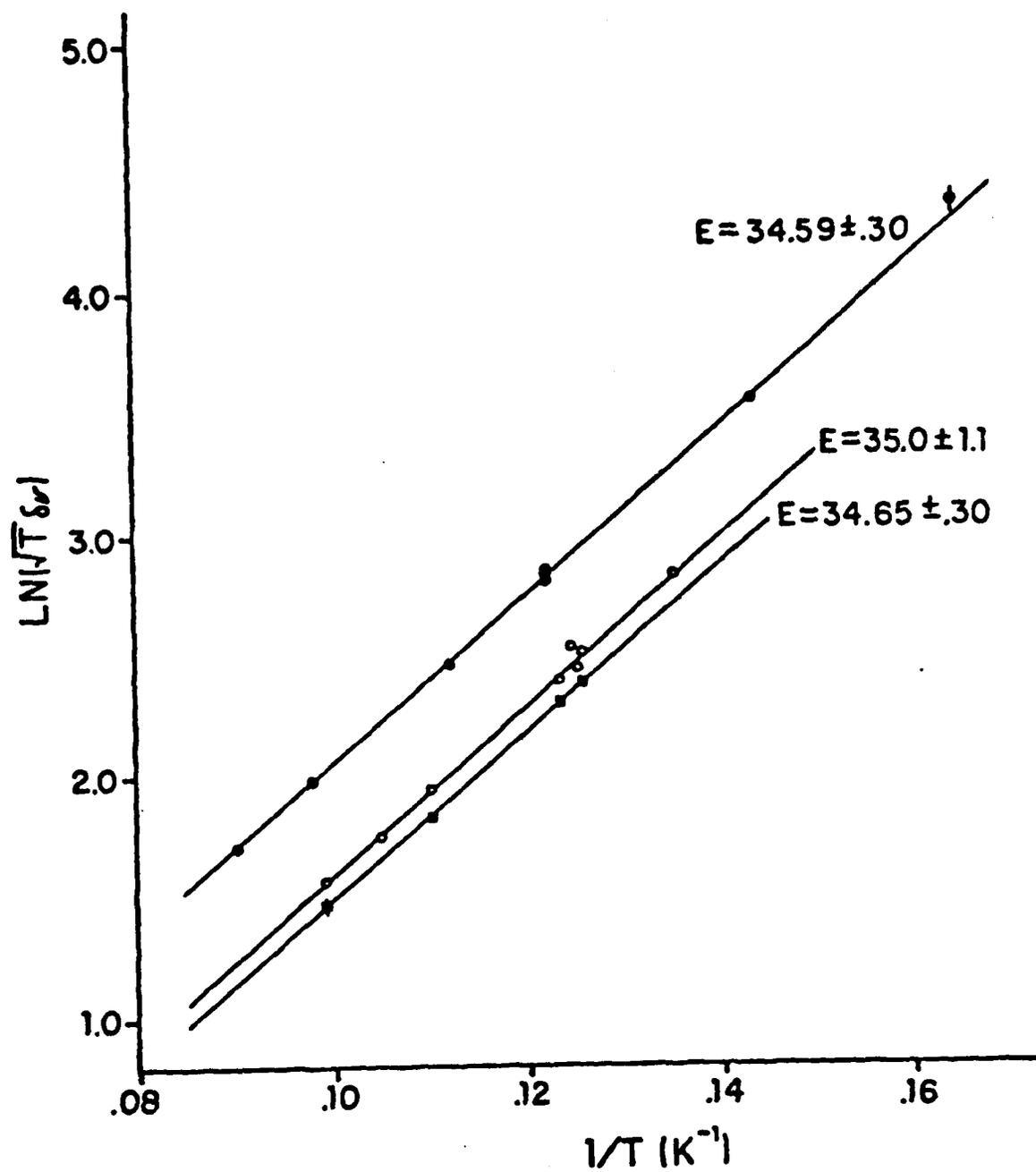


FIGURE 6 SOLID ATOMIC NEON SURFACE WALL SHIFTS

ACKNOWLEDGEMENTS

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