HYDROGEN MASER OSCILLATION AT 10 K

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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 hydrogen[1], 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[5] 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 transiton 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

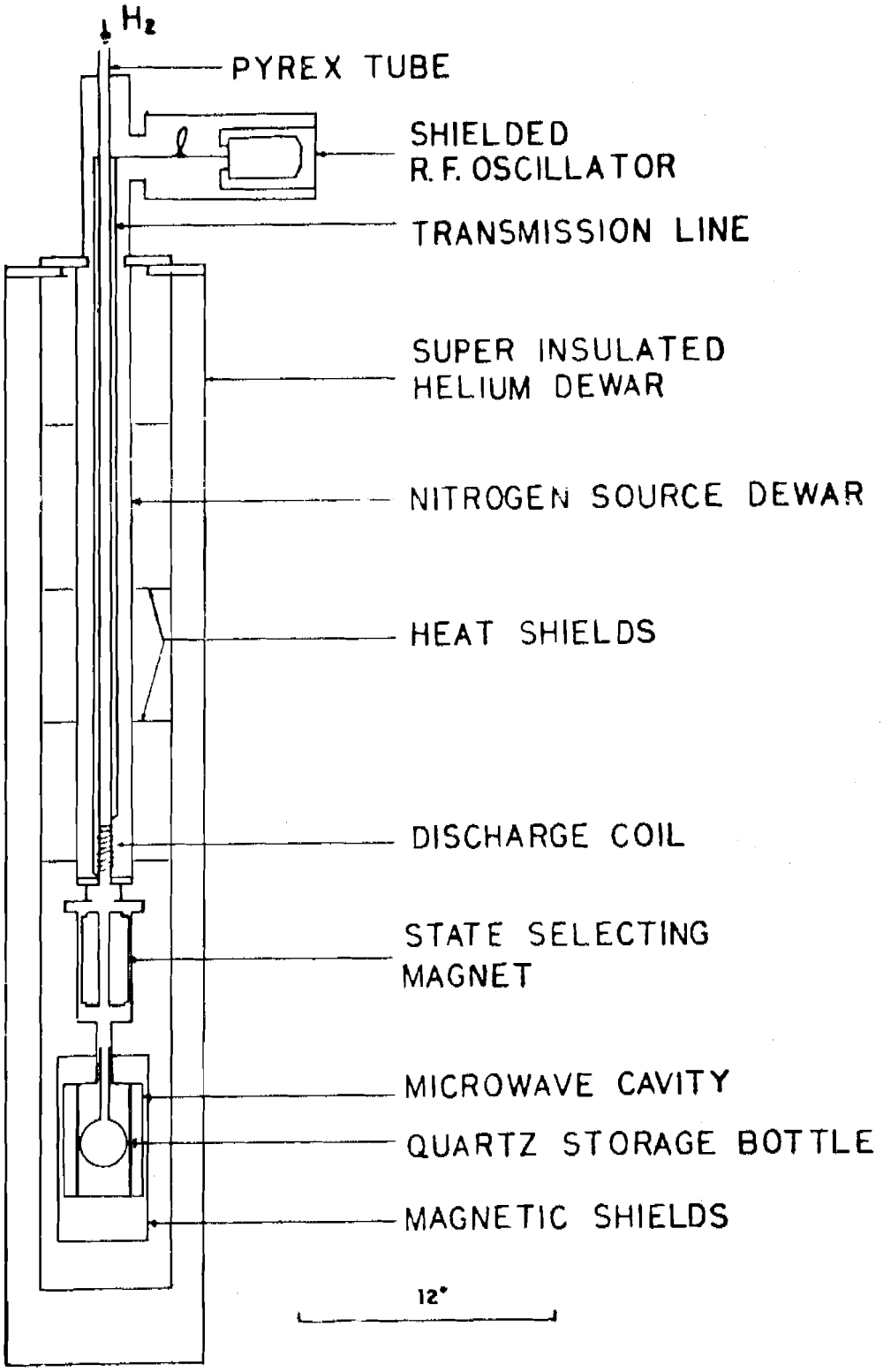

FIGURE 1 SCHEMATIC OF THE APPARATUS

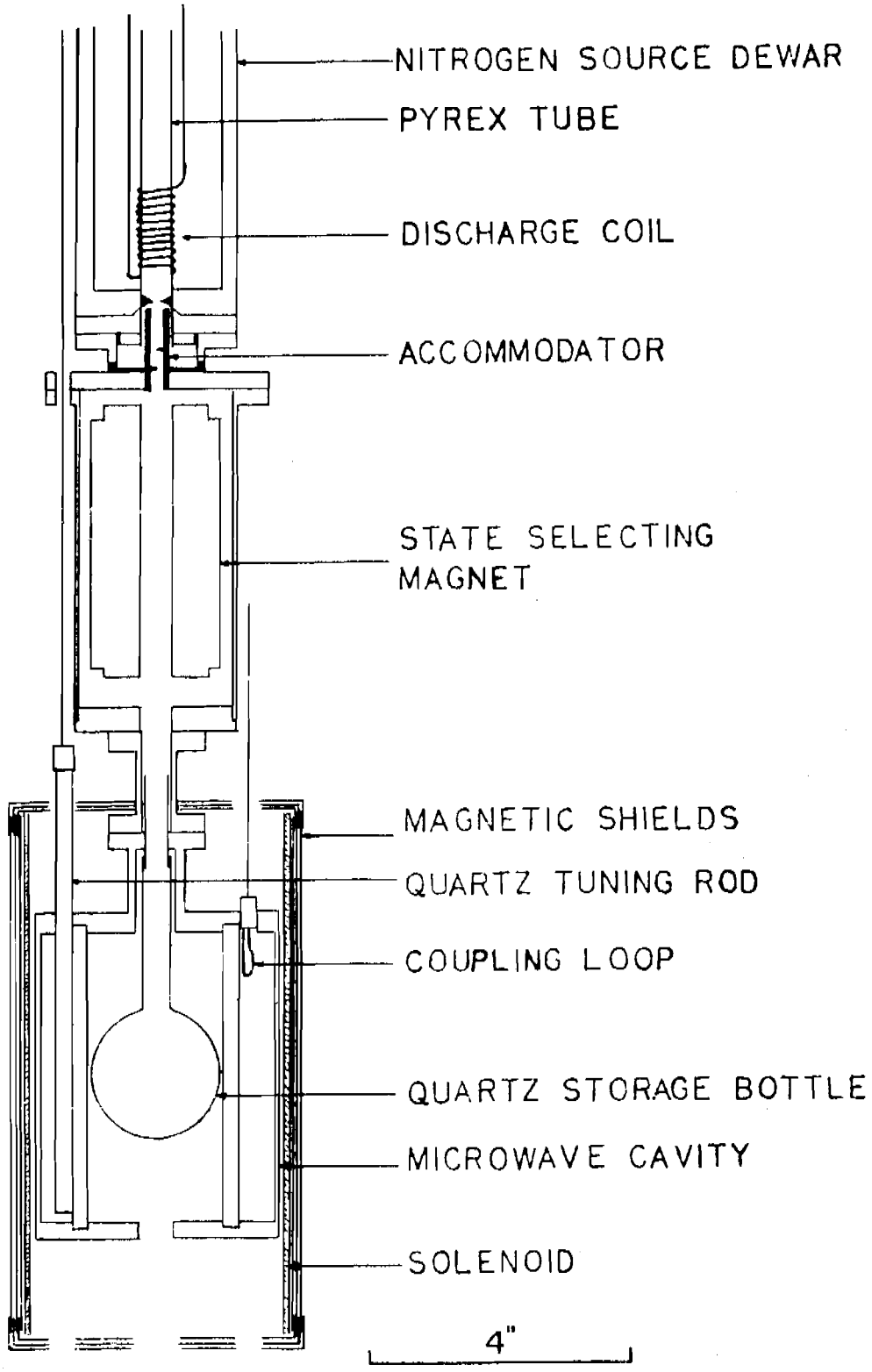

FIGURE 2 SCHEMATIC OF HYDROGEN SOURCE AND CAVITY ASSEMBLY

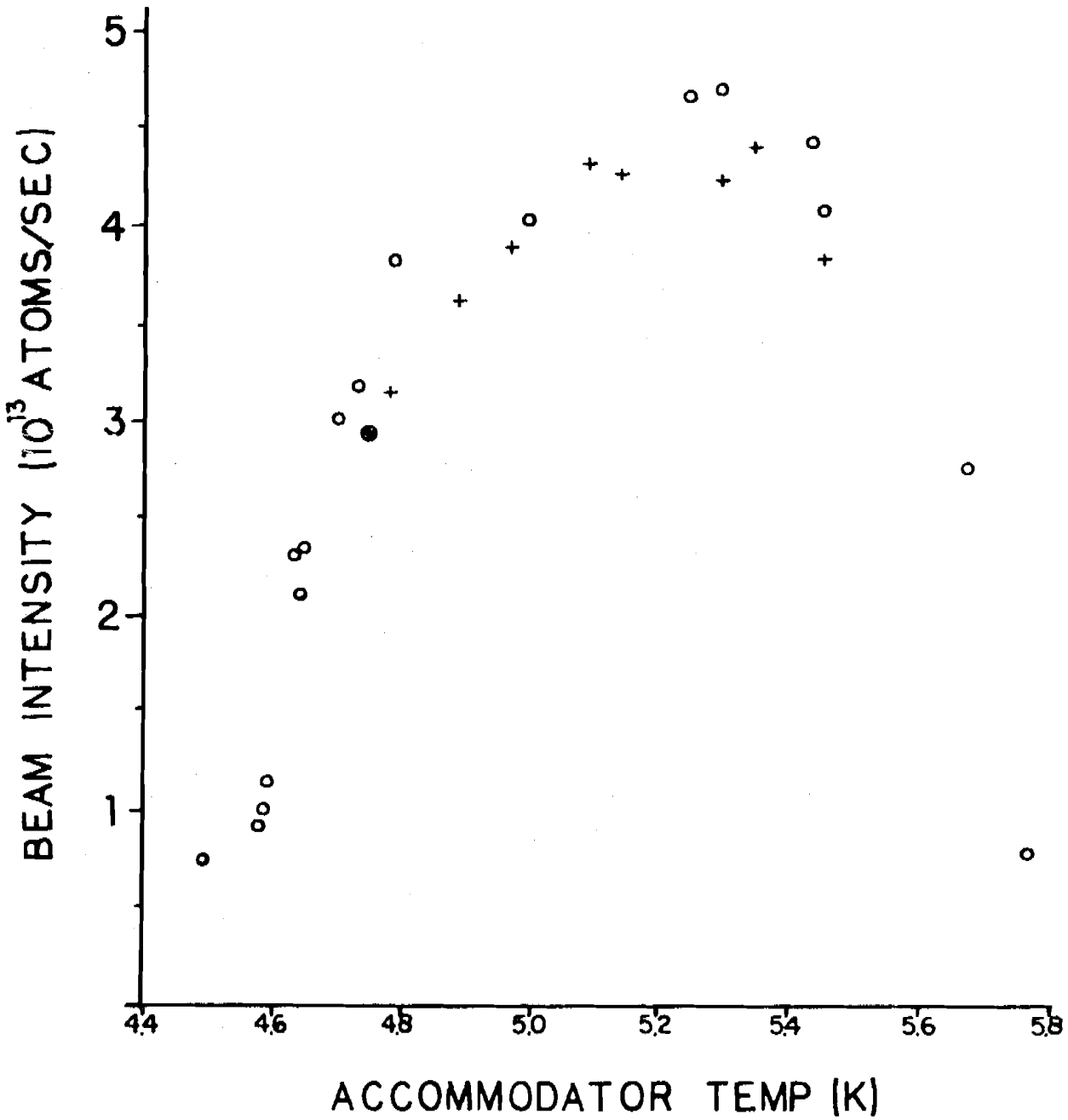

FIGURE 3 FLUX OF F=1, m_F=0 HYDROGEN ATOMS

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 H_2 at the somewhat lower temperatures in the earlier work. Theoretical calculations[7] of the binding of H to solid H_2 indicate that there should be a single bound state for motion perpendicular to the surface and almost complete freedom to move laterally. Theory[6] 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 H_2 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 H_2 vapor pressure prevents that.

Relaxation processses 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 H_2: 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 H_2 vapor pressure at temperatures where the wall shifts and relaxation rates would be low.

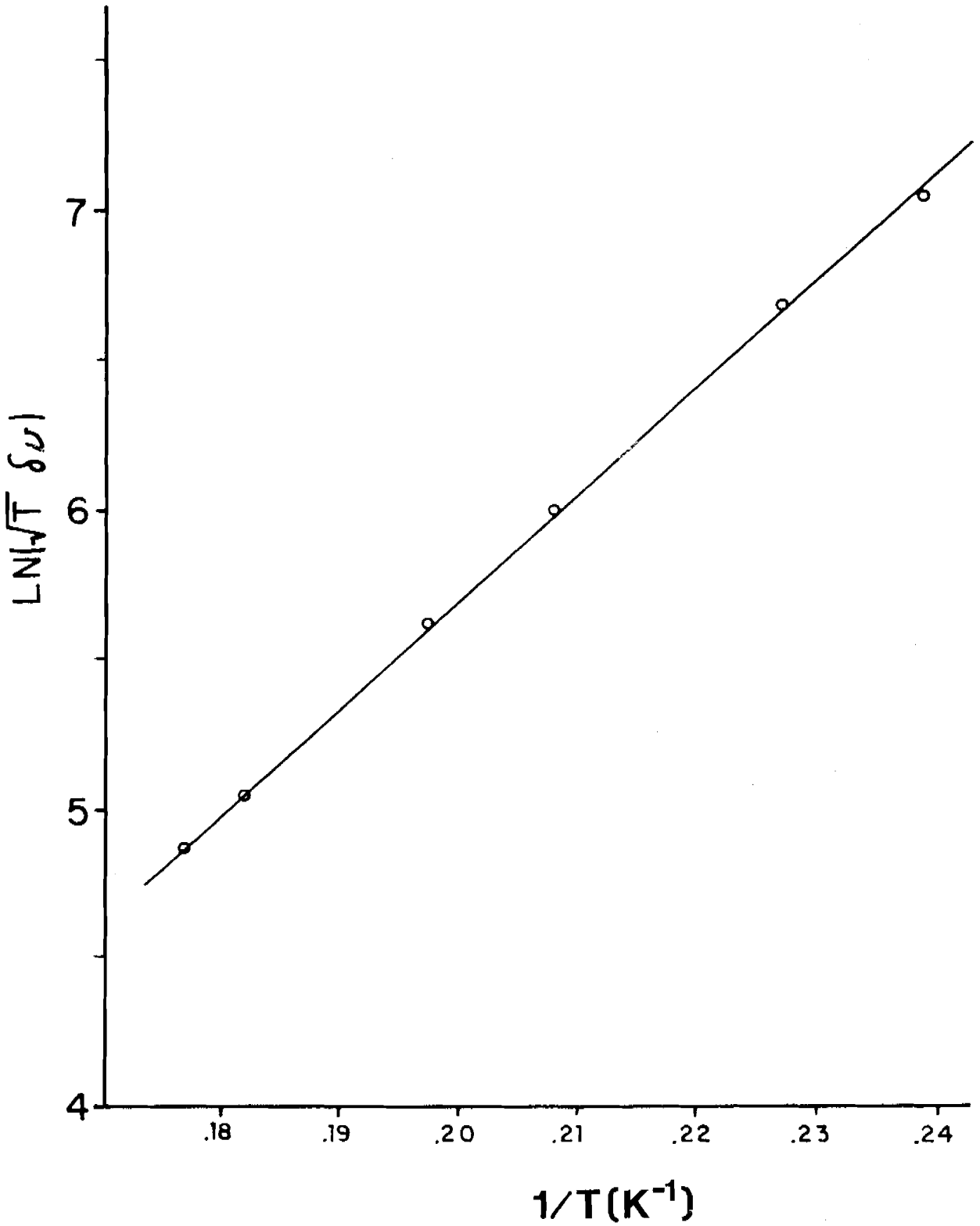

FIGURE 4 MOLECULAR HYDROGEN SURFACE WALL SHIFTS

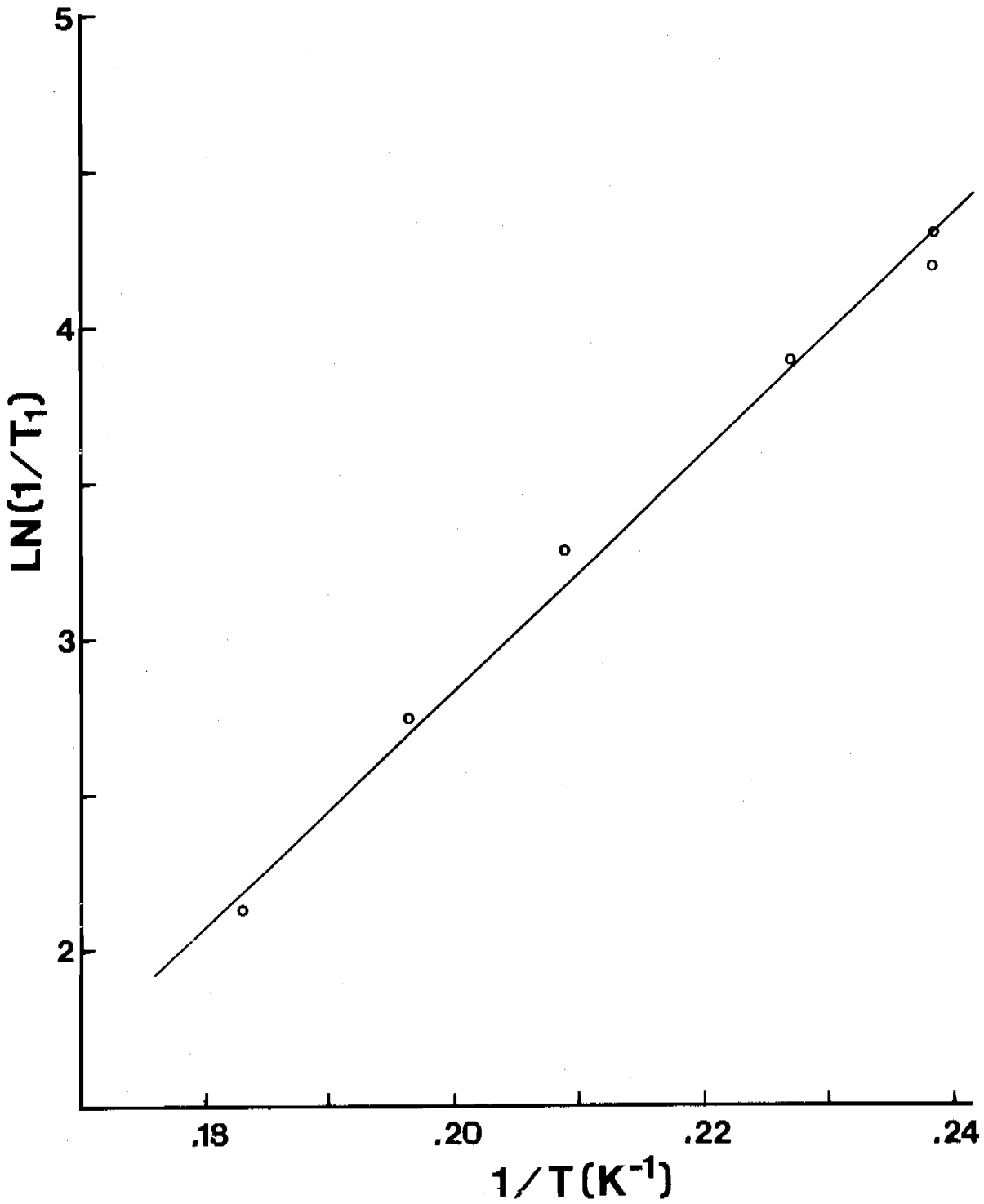

FIGURE 5 MOLECULAR HYDROGEN SURFACE RELAXATION RATES

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 predicted[7] 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 surafce 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.

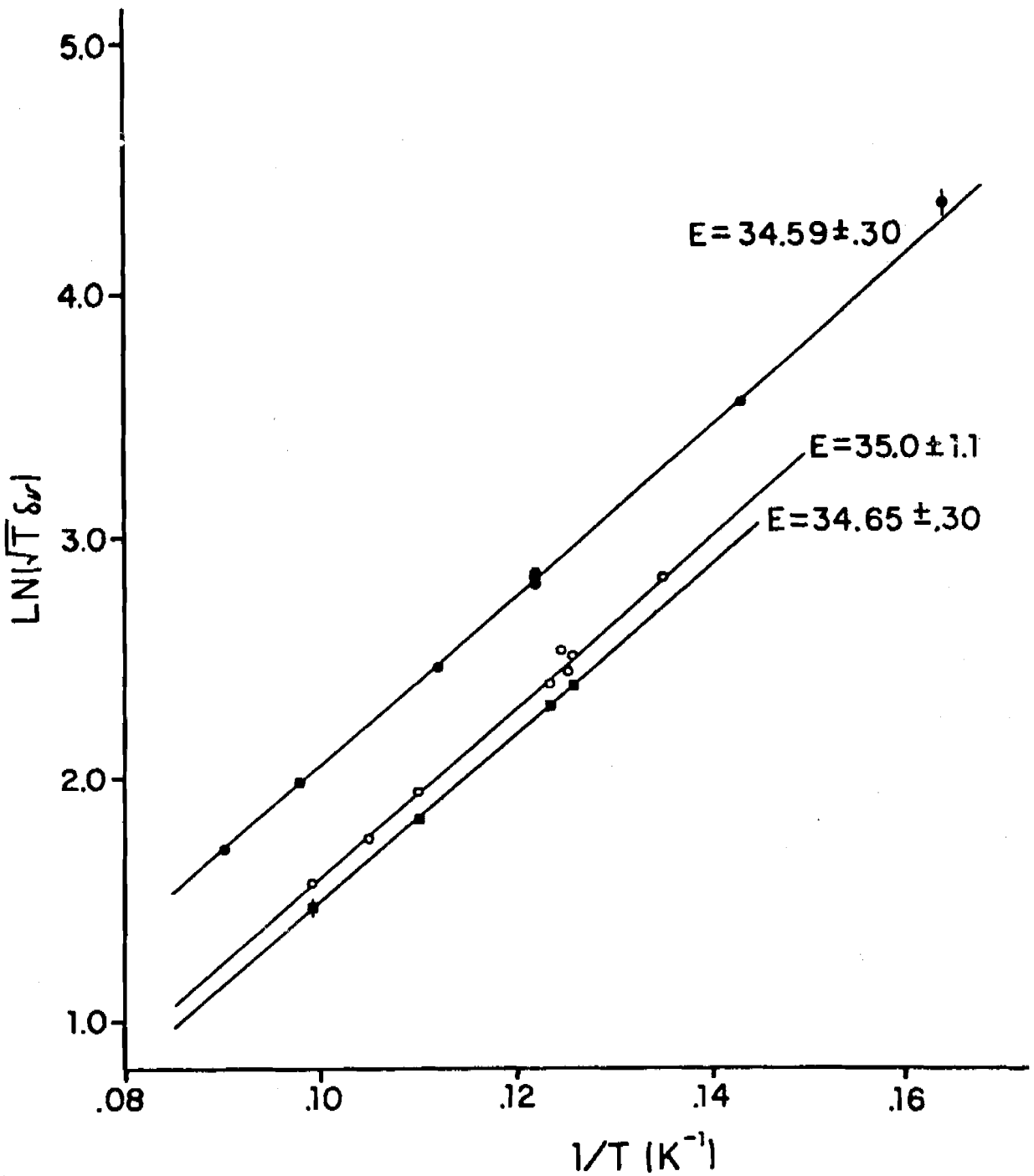

FIGURE 6 SOLID ATOMIC NEON SURFACE WALL SHIFTS

ACKNOWLEDGEMENTS

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