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HYPERFINE CONTRIBUTION TO SPIN-EXCHANGE FREQUENCY SHIFTS IN THE HYDROGEN MASER*

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Summary

We have recalculated shifts of the ground state $\Delta m_F=0$ transition of a gas of hydrogen atoms in low magnetic field due to electron spin-exchange collisions between the atoms. We predict a new source of frequency shifts not compensated for by the usual methods of tuning the microwave cavities of oscillating hydrogen maser frequency standards. Near room temperature these additional shifts are small, but large enough to affect stability at the level of $\delta\omega/\omega \approx 10^{-15}$. At very low temperatures these shifts are large compared to the potential thermal instabilities of cryogenic hydrogen maser standards. Above 5 Kelvin they decrease rapidly and so are less severe when using neon storage surfaces near 10 K than when using liquid helium storage surfaces near 0.5 K.

Introduction

Collisions between the hydrogen atoms radiating in a hydrogen maser frequency standard affect the maser frequency in two important ways. They directly shift the transition frequency, and they broaden the radiative linewidth, which increases the frequency pulling due to cavity mistuning. The usual theoretical treatment of hydrogen atom spin-exchange collisions, which treats the energy levels during the collisions as degenerate, predicts that the direct spin-exchange frequency shifts have the same dependence on radiative linewidth as frequency shifts due to cavity mistuning.[1] Tuning the cavity so that the oscillation frequency is independent of radiative linewidth is predicted by that treatment to leave the oscillation frequency independent of collision rate.[2] Such "spin exchange tuning" methods have been important to the development of hydrogen maser standards because drifting hydrogen atom beam intensities induce changing collision rates.

Including the hyperfine structure of the energy levels during collisions to first order, but assuming undeflected classical collision trajectories and ignoring the identity of the colliding atoms, predicts additional direct frequency shifts which leave the spin exchange tuned oscillation frequency offset by an amount proportional to that part of the radiative linewidth not caused by collisions.[3] Measurements of this offset in a room temperature hydrogen maser confirmed within errors a numerical estimate of this effect.[3] The offset predicted by this calculation does not affect the stability of hydrogen maser standards unless something happens to affect that part of the linewidth not due to collisions, such as a change of relaxation by motion of the atoms through magnetic field gradients or a change of relaxation due to interactions with the storage surface.

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We have recently recalculated the additional direct shifts quantum mechanically, including atom identity.[4] We find new effects which are nonlinear in the collision rate and so produce not only an offset of the spin exchange tuned oscillation frequency, but also a variation of the oscillation frequency with collision rate even after spin exchange tuning. Theoretical details can be found in the recent paper by Verhaar, Koelman, Stoof, Luiten and Crampton, hereafter referred to as (VKSLC).[4] Here we present only the results and then illustrate their implications for hydrogen maser frequency standards operating at room temperature and at cryogenic temperatures.

Spin-Exchange Frequency Shifts

Shifts of the hydrogen maser oscillation frequency due to collisions between the atoms, including direct shifts and shifts dependent on the radiative linewidth, are given by VKSLC Eq. (17) as

$$\delta\omega = [\Delta+\alpha\bar{\lambda}_0(1+\Delta^2)-\Omega]/\tau_2 + \Omega/\tau_0 \qquad (1)$$

with Δ the ratio of cavity mistuning to cavity width, α a constant dependent on cavity Q and filling factor, $\bar{\lambda}_0$ the cross section for frequency shifts proportional to the transition level population difference, τ_2 the radiative lifetime (inversely proportional to the linewidth $1/\pi\tau_2$), and τ_0 the partial lifetime without relaxation due to collisions. Ω is

$$\Omega = -\frac{\bar{\lambda}_1(\rho_{cc}+\rho_{aa})+\bar{\lambda}_2}{\bar{\sigma}_1(\rho_{cc}+\rho_{aa})+\bar{\sigma}_2} \qquad (2)$$

with $\bar{\lambda}_1$ and $\bar{\sigma}_1$ the cross sections for frequency shifts and broadening proportional to the sum $(\rho_{cc}+\rho_{aa})$ of level populations involved in the transition and $\bar{\lambda}_2$ and $\bar{\sigma}_2$ the cross sections for frequency shifts and broadening not proportional to level populations.[4] The cross sections are plotted in Fig. 1, and $\alpha\bar{\lambda}_0$ and Ω are plotted in Fig. 2 for particular choices of maser parameters and $(\rho_{cc}+\rho_{aa})$.

Ω generally depends on collision rate in a complicated way via the collision rate dependence of the level population sum $(\rho_{cc}+\rho_{aa})$. Careful measurements of oscillation frequencies, level populations and cavity mistunings can in principle determine the cross sections and the interesting physics which underlies them. However, the important questions for hydrogen maser standards are (1) whether these effects lead to important new sources of frequency instabilities because they couple the oscillation frequency to relaxations due to collisions between the atoms and to other relaxation mechanisms, and (2) whether there are strategies for minimizing these new sources of frequency instabilities.

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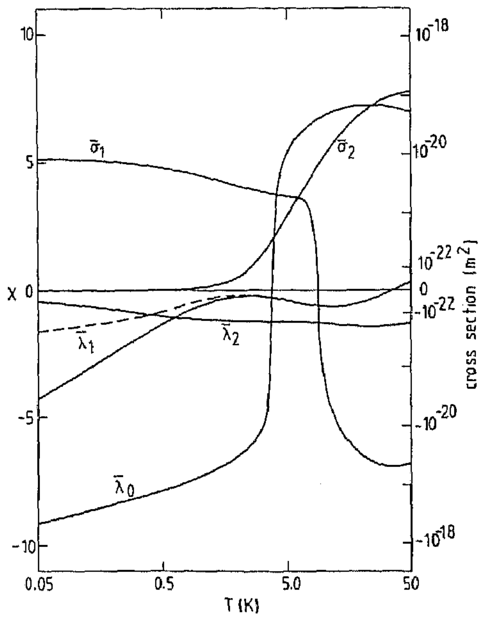

Fig. 1 Thermally averaged values of the various frequency shift and broadening cross sections as functions of absolute temperature. The left hand vertical scale is linear in $\chi = \mathrm{arcsinh}(\bar{\sigma}_i/10^{-22}\ m^2)$ and $\chi = \mathrm{arcsinh}(\bar{\lambda}_i/10^{-22}\ m^2)$. Solid lines, calculation to all orders; dashed line, first order calculation where it differs significantly from the calculation to all orders.

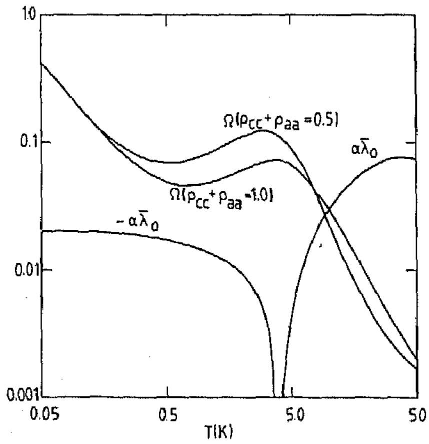

Fig. 2 The dimensionless frequency offset parameters $\alpha\bar{\lambda}_0$ and Ω as functions of absolute temperature. Ω is given for $\rho_{cc}+\rho_{aa}$ assumed constant at 0.5 and 1.0, and $\alpha\bar{\lambda}_0$ is given for a typical choice of cavity Q and filling factor.

Room Temperature Maser Standards

We illustrate these effects with simulations of a maser near room temperature having the operating parameters of the maser used in the experiment reported in Ref. 3 and our own preliminary values for the collision cross sections at 300 degrees Kelvin. We ignore relaxations due to magnetic field gradients and collisions with the surfaces, and we use rate equations for the evolution of level populations due to collisions assuming degenerate states during collisions.[5] Fig. 3 displays a simulation of the oscillation power level and oscillation frequency as the hydrogen atom collision rate is varied by varying the input beam intensity. $1/\tau_2$ provides a convenient measure of relaxation rates, including here only atom flow and collisions, because it can be determined from changes of $\delta\omega$ with Δ, as shown by Eq. (1) above.

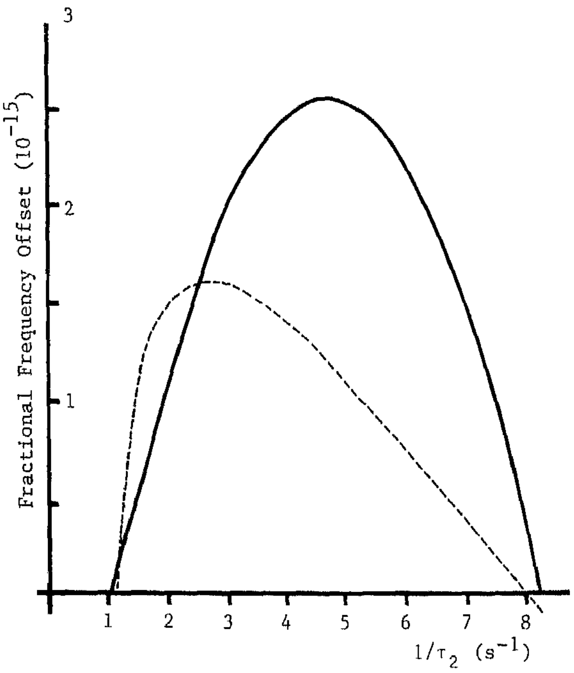

Fig. 3 Solid line: oscillation power level (arbitrary units) plotted against $1/\tau_2$. Self- sustained maser oscillation is obtained for $1/\tau_2$ ranging from 1.16 s^{-1} to 8.18 s^{-1}. Dashed curve: variations of fractional oscillation frequency offset $\delta\omega/\omega$ (from a base offset $\delta\omega/\omega = 5.5\times10^{-14}$) with $1/\tau_2$ over the full range of oscillation.

The Fig. 3 variation of oscillation frequency with $1/\tau_2$ is for a case when the cavity has been tuned so that the oscillation frequency is the same at the minimum and maximum collision rates at which self-sustained oscillation can be obtained. The fractional frequency offset at those two end points is $\delta\omega/\omega = 5.49 \times 10^{-14}$, and the offset varies nonlinearly between those two points because of the τ_2 dependence of Ω. Using a large range of τ_2 variations to set the cavity tuning does minimize the uncertainty of cavity tuning due to thermal fluctuations of the oscillation frequency at the two collision rates chosen as tuning points,[2] but it leaves a fractional variation of frequency with collision rate of order 10^{-15} per Hz of radiative linewidth at high collision rates and an even steeper variation of frequency with collision rate at collision rates just above the threshold for oscillation. In addition, the overall fractional frequency offset varies with $1/\tau_0$, so that a change of τ_0 due to a change of some relaxation rate produces a fractional frequency change at the 10^{-15} level, in addition to changes of any direct frequency shifts caused by those relaxations.[6] The effects due to $\bar{\lambda}_1$ and $\bar{\lambda}_2$ are just large enough to be significant for hydrogen maser standards under some conditions, but they are difficult to detect because of the long averaging times required to make measurements at the 10^{-14} to 10^{-15} level with room temperature masers. Alternatively, there may be strategies that will minimize these effects. For example, Fig. 4 displays the variations of oscillation power and frequency for the same parameters except that a higher collision rate has been chosen for the lower of the two tuning point collision rates. The precision of the cavity tuning using the tuning points displayed in Fig. 4 is less by about 1/4, but the variation of oscillation frequency with collision rate is only of order 10^{-16} if the collision rates are restricted to values between the two tuning points. Alternatively, the overall frequency offset might be reduced by tuning the cavity so that the oscillation frequency is independent of τ_0, but at the cost of greatly increasing the dependence of oscillation frequency on collision rate.

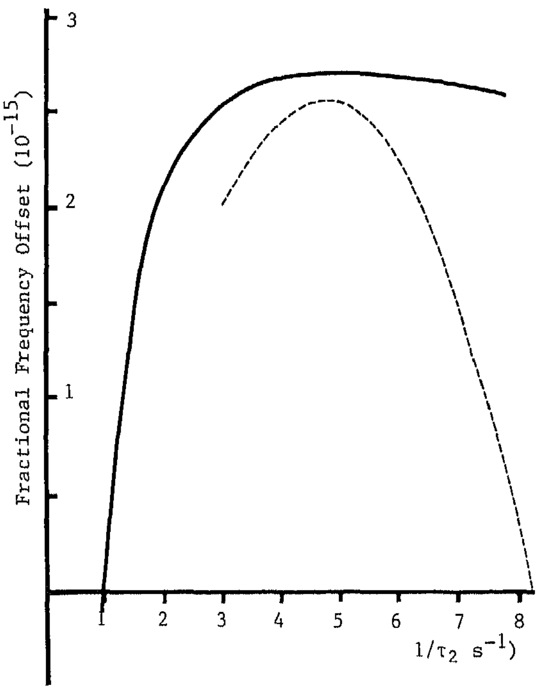

Fig. 4 Dashed curve: oscillation power level (arbitrary units) plotted against $1/\tau_2$ only for values of $1/\tau_2$ between the two collision rates used for tuning. Solid curve: variations of fractional oscillation frequency offset $\delta\omega/\omega$ (from base offset $\delta\omega/\omega$ = 5.5 x 10^{-14}) with $1/\tau_2$ over the full range of values for which there is oscillation.

Adding additional relaxations due to magnetic field gradients and collisions with the storage surfaces does not affect the results qualitatively. Overall fractional frequency offsets remain of order 10^{-14}, and deviations of fractional frequency offsets with collision rate remain of order 10^{-15}, but these can be reduced significantly by using smaller ranges of collision rates for cavity tuning.

Liquid Helium Temperature Maser Standards

Although the three liquid helium storage surface hydrogen masers developed to date[7-9] have not achieved numbers of radiating atoms as high as those in room temperature standards, higher numbers of atoms are essential to achieving the improvements of short term frequency stability potentially possible because of the decrease of spin exchange relaxation cross sections with decreasing temperature[10-12]. We assume that standards having atom densities high enough to produce large collision contributions to $1/\tau_2$ will be developed. Fig. 5 displays the results of a simulation assuming atom flow rate 1 s^{-1} and maximum collision rates high enough to increase $1/\tau_2$ to 8 s^{-1}.

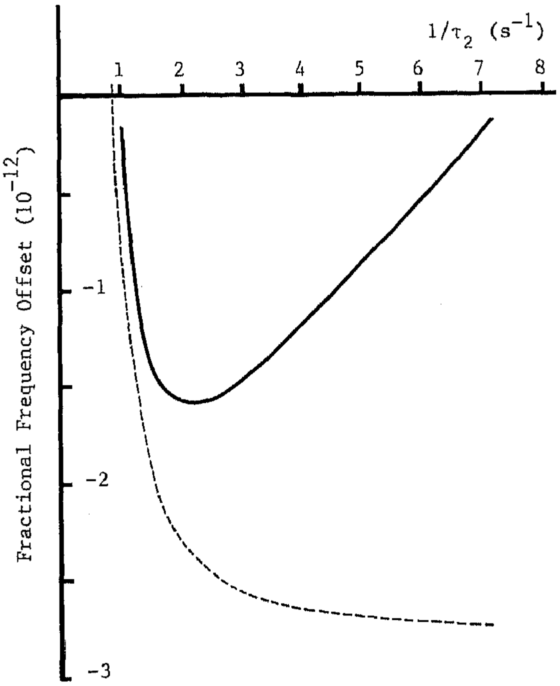

Fig. 5 Dashed curve: variations of fractional oscillation frequency offset $\delta\omega/\omega$ (from a base offset -5.5 x 10^{-12}) with $1/\tau_2$ when the maser has been tuned for equal oscillation frequencies at the minimum collision rate for oscillation and the maximum collision rate available. Solid curve: variations of fractional oscillation frequency offset when the maser is tuned using a higher collision rate for the lower of the two tuning point collision rates.

The Fig. 5 behavior is similar to that in Fig. 3 and Fig. 4, except that the sign of the frequency offset is inverted and the scale is three orders of magnitude larger. The problems posed by these new sources of frequency shifts are most severe if only a small variation of $1/\tau_2$ can be made by varying the collision rate. The problems are much less severe if collision rates dominate $1/\tau_2$, but they are still very large when compared to the potential thermal instabilities of cryogenic masers and even when compared to the stabilities of current room temperature standards. If liquid helium surface hydrogen masers are to be competitive as frequency standards, design studies must take into account these new sources of frequency shifts.

Middle Ground: Neon Surface Masers

Similar simulations using the parameters of neon surface hydrogen masers operating near 10 Kelvin reveal behavior that is qualitatively the same as the 0.5 K behavior, except that the scale is less by a factor 25 because of the decrease of the spin-exchange frequency shift cross sections with increasing temperature. However, the spin-exchange relaxation cross sections are still low enough that collisions are not likely to limit the radiated power at achievable hydrogen atom beam intensities. If solid neon surfaces that are as stable as liquid helium surfaces can be developed, the neon surface hydrogen masers offer a technologically attractive alternative to the liquid helium surface masers as frequency standards.

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