

## Hyperfine Contribution to Spin-Exchange Frequency Shifts in Cryogenic Hydrogen Masers

B.J. VERHAAR, J.M.V.A. KOELMAN, H.T.C. STOOF, O.J. LUITEN and S.B. CRAMPTON\*

Department of Physics, Eindhoven University of Technology,  
 5600 MB Eindhoven, The Netherlands\*\*

We have rigorously included hyperfine interactions during electron-spin-exchange collisions between ground state hydrogen atoms. We predict additional frequency shifts which are not compensated for by the usual methods of tuning maser cavities. These shifts are large compared to the potential thermal instabilities of cryogenic masers and are very sensitive to details of the interactions, especially the treatment of non-adiabatic effects.

### 1. INTRODUCTION

In the usual (degenerate-internal-states) treatment of electron-spin-exchange collisions between paramagnetic atoms [1-5] hyperfine interactions are ignored during the collisions. These calculations predict a small shift of the frequency of ground state hydrogen atoms which collide while radiating on the low magnetic field  $\Delta m_F=0$  ( $c \leftrightarrow a$ ) hyperfine transition (Fig. 1). This frequency shift is proportional to the difference of the  $c$ - and  $a$ -state atom densities, and that level density difference is itself proportional to the full atomic resonance linewidth when there is self-excited maser oscillation [6], so that this spin-exchange frequency shift is compensated for by cavity pulling in atomic hydrogen masers oscillating on the  $\Delta m_F=0$  transition [7]. Including hyperfine interactions during the collisions semi-classically to first order without including the effects of atom identity reveals additional shifts proportional to the durations of the collisions and not compensated for by cavity pulling [8]. We present here the results of a fully quantum mechanical treatment, including both atom identity and hyperfine-induced effects to all orders, which predicts hyperfine-induced frequency shifts having a more complicated dependence on atomic level populations than the predictions of the semi-classical calculation. At low temperatures the hyperfine induced shifts are large and therefore significant to the operation of neon surface [9] and liquid helium surface [10-12] hydrogen masers.

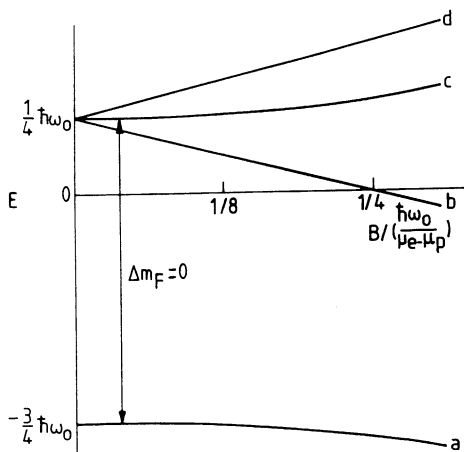


Fig. 1: Energy levels of the hydrogen hyperfine states as a function of magnetic field.

### 2. RESULTS

Symmetry considerations reduce the problem to determining elastic S-matrix elements for a few pairs of hyperfine levels of the two colliding atoms as they evolve through coupled channels during the collision. The method and details of the calculations are presented elsewhere [13-15]. Here we present only the results.

We find the usual shift proportional to difference of  $c$  and  $a$  state atom densities  $n_c - n_a$  and a hyperfine-induced frequency shift proportional to the total atom density  $n_H$ , as in the semi-classical treatment, but also a hyperfine-induced shift proportional to the sum of the  $c$  and  $a$  state atom densities  $n_c + n_a$ :

$$\delta\omega = (n_c - n_a)v_{th}\lambda_0 + (n_c + n_a)v_{th}\lambda_1 + n_H v_{th}\lambda_2$$

where  $v_{th}$  is the thermally averaged collision speed and the  $\lambda_i$  quantities are thermally averaged effective cross sections. In Fig. 2 the  $\lambda$  "cross sections" are plotted as functions of temperature.

### 3. SIGNIFICANCE TO HYDROGEN MASERS

Although  $\lambda_1$  and  $\lambda_2$  are in general small compared to  $\lambda_0$ , in an oscillating H-maser  $(n_c + n_a)\lambda_1$  and  $n_H\lambda_2$  are certainly not small compared to  $(n_c - n_a)\lambda_0$  because  $n_c - n_a$  is sharply reduced by transfer of energy to the cavity electromagnetic field. For self-excited maser oscillation in which the atoms couple to a single Lorentzian microwave cavity mode having center frequency  $\omega_c$  and width  $\omega_c/Q$ , the  $\Delta m_F=0$  level density difference is [6]

$$n_c - n_a = \gamma(1 + \Delta^2)\tau_2^{-1}$$

with  $\Delta = Q(\omega_c/\omega - \omega/\omega_c)$ ,  $1/\pi\tau_2$  the full frequency width (in Hz) of the  $\Delta m_F=0$  transition, and  $\gamma$  a constant of order  $10^9 \text{ cm}^{-3}\text{s}$ . Substituting this density difference and including the effects of cavity mistuning, the collisional frequency shift becomes

$$\delta\omega = [\Delta + \gamma v_{th}\lambda_0(1 + \Delta^2)]\tau_2^{-1} + (n_c + n_a)v_{th}\lambda_1 + n_H v_{th}\lambda_2$$

\*Department of Physics and Astronomy, Williams College, Williamstown, MA 01267, USA

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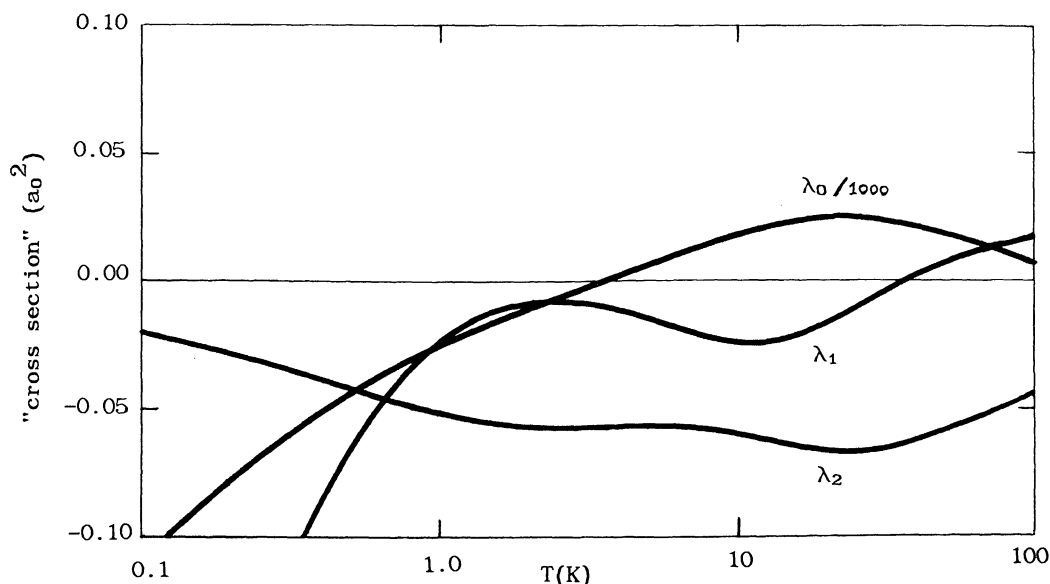


Fig. 2 : The thermally averaged frequency shift "cross sections" as functions of temperature.

Using the above value of  $\gamma$  we find  $\gamma v_{th} \lambda_0 \approx 0.02$  at subkelvin temperatures, yielding a shift by about 2% of the resonance linewidth. However, this shift has the same dependence on relaxation processes as the cavity pulling and can be compensated for by adjusting  $\Delta$ . The hyperfine induced shifts, proportional to  $n_c + n_a$  and  $n_H$  do not depend on relaxation processes through a simple factor  $1/\tau_2$  and so cannot be compensated for by cavity pulling. At atom densities high enough to achieve maser oscillation with good stability, collisions contribute appreciably to the linewidth, so that rough estimates of the hyperfine-induced frequency shifts relative to the full resonance linewidth can be made by comparing the hyperfine-induced lineshifts to the collisional line broadening. For low temperatures this ratio becomes very large and for  $T \rightarrow 0$  it even diverges as  $1/\sqrt{T}$ . In the temperature range  $0.2 \text{ K} < T < 7 \text{ K}$  the hyperfine induced shifts are larger than 6% of the collisional linewidth. At temperatures above 7 K the hyperfine-induced frequency shift rapidly diminishes. At liquid nitrogen temperature we find a hyperfine induced shift equal to 0.1% of the collisional linewidth.

#### CONCLUSION

Including hyperfine interactions during collisions yields additional frequency shifts not compensated for by the usual method of tuning maser cavities, and which amount to several percent of the collisional linewidth at the operating temperatures of  $\ell$ - $^4\text{He}$  coated cryogenic H-masers. Such large values put severe limits on maser parameter stabilities required to achieve long term maser frequency instabilities as low as the thermal instabilities predicted [16] for cryogenic H-masers.

On the other hand, their sensitivity to hyperfine-induced frequency shifts should usefully test the details of these calculations, including the non-adiabatic corrections to the continuum states.

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