Notes on Cryomaser Spin-Exchange

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January 29, 2002

1 Introduction

Let's try to understand how we're going to extract Verhaar style spin-exchange parameters from the cryomaser experiment. Marc handed me a bunch of papers to use as reference. He has me start with section 3.C of Ron's thesis [1] along with a page from a grant proposal that is very similar. Then he has me reading work of the UBC guys. They have both a nice short paper [2] and a much more detailed phys. rev. A [3]. To make sure that I remember how to measure q and γ_t he also gave me a copy of Ed's paper on such things [4].

I also grabbed Crampton's paper on generalizing the spin-exchange formulation [5] and a slightly earlier conference proceedings [6] both for the introduction to spin-exchange and for discussions of Magnetic Inhomogeneity Shifts (MIS). Also, it's useful to look at Ron's room temperature measurements [7] and the pre-bulb status report on the cryomaser [8].

2 Spin-Exchange equations

The spin-exchange parameter we are trying to extract is Ω which shifts the maser frequency, ω as

$$\omega - \omega_0 = \left[\Delta + \alpha\bar{\lambda}_{se}\right]\gamma_2 - \Omega\gamma_H \tag{1}$$

where ω_0 is the atomic frequency. $\Delta = (2Q_c/\omega_0)(\omega_c - \omega_0)$ is the "cavity-pulling" parameter where Q_c is the loaded cavity quality factor and ω_c is the cavity frequency. γ_2 is the full atomic linewidth while γ_H is the portion of the linewidth resulting from hydrogen-hydrogen collisions. $\bar{\lambda}_{se}$ is the degenerate internal states spin-exchange cross section and α is a coefficient proportional to the mean thermal velocity of the hydrogen as well as various system constants.

Ω can be expressed in terms of Verhaar's parameters as

$$\Omega = -\frac{\bar{\lambda_1}\left(\rho_{cc} + \rho_{aa}\right) + \bar{\lambda_2}}{\bar{\sigma_1}\left(\rho_{cc} + \rho_{aa}\right) + \bar{\sigma_2}} \tag{2}$$

where the λ's are frequency shift parameters and the σ's are line broadening parameters. $\rho_a a$ is the population of the lower masing state and $\rho_c c$ is the population in the upper masing state. Thus, their sum is just the population participating in maser action. In the cryomaser with its simple hexapole state-selector should ideally have $\rho_{aa} + \rho_{cc} \approx 1/2$ as half the population entering the maser is ρ_{cc} and half is ρ_{dd}.

3 Marc's proposal

Marc, following Ron's proposal and the analysis of the UBC guys basically say that we should measure the maser frequency and power as a function of cavity detuning and flux. What we'd like to do is to plot changes in maser frequency as a function of γ_H to extract Ω. The first step in this process will be to determine γ_H.

We can determine the total linewidth from a "line-Q" (Q_l) measurement. We measure the maser frequency as a function of cavity detuning. From equation (1) we see that the slope of this plot should be

$$\frac{\partial \omega}{\partial \omega_c} = \frac{\omega_0}{2Q_c}\pi\gamma_2 \tag{3}$$

in which all the parameters can be measured and γ_2 extracted.

The next step is to determine the density independent part of the linewidth which is really just $\gamma_2 - \gamma_H$. We can determine this by varying the flux and plotting Q_l as a function of power. From Ed's analysis [4] (eq. 10) we see that the dependence of Q_l on power is

$$Q_l^{-1} = \frac{2}{\omega}\left(\gamma_d + \gamma_2'\right)\left[1 + \frac{q}{1-cq}\frac{\gamma_t}{\gamma_d + \gamma_2'}\right] + A\frac{q}{1-cq}\frac{1}{\gamma_t}\ P \tag{4}$$

where

$$A = \frac{16\pi\mu_0^2 Q_c \eta}{\omega^2 \hbar^2 V_c}. \tag{5}$$

γ_d is the rate at which atoms leave the bulb either through geometric escape or through recombination. γ_1' and γ_2' are the polarization and coherence loss mechanisms which are not dependent on density but are in addition to atomic loss. We expect these to be primarily magnetic losses. γ_t is a somewhat mysterious combination of these elements which is defined as

$$\gamma_t^2 = \left(\gamma_d + \gamma_1'\right)\left(\gamma_d + \gamma_2'\right) \tag{6}$$

where all the rates are the same as above. If we accept the Ed ansatz that $\gamma_1' = \gamma_2'$ then $\gamma_t = \gamma_2 - \gamma_H$ and we have found the spin-exchange free component of the linewidth. If we return to equation (4) and maintain Ed's ansantz, we will find that γ_t can be extracted from a linear fit to the Q_l measured versus maser power.

There is a slight hitch in this line of reasoning, however. That is that A in equation (4) (which is defined in eq. (5)) relies on knowledge of the volume of the cavity and the filling factor. We can probably determine these to an accuracy sufficient to our needs, but we don't currently really know what they are. The UBC analysis [3] is much more heroic in this regard: they fill the maser volume with liquid oxygen and look at how it loads the cavity in an attempt to accurately determine the filling factor!

Thus, Marc's proposal begins with the 2 dimensional grid of frequencies and powers ($\Delta\omega$, P) as a function of detuning and flux ($\Delta\omega_c$, I). At a fixed flux and thus power, we determine Q_l by fitting $\Delta\omega$ vs. $\Delta\omega_c$. Then at varying fluxes, we can plot Q_l vs. P. This plot should be linear, and from the fit parameters, we should be able to extract γ_t, which following the ansatz, is the spin-exchange independent linewidth.

Now having determined the spin-exchange independent linewidth, we can plot Q_l vs. maser power and use this to determine at fixed detuning, $\Delta\omega$ vs. $(\gamma_2 - \gamma_t)$ where $\gamma_2 = \omega/(2Q_l)$. Rewriting eq. (1) in terms of γ_2 and γ_t we find that

$$\Delta\omega = \left[\Delta_c + \alpha\bar{\lambda}_{se}(1+\Delta_c^2)\right]\left(\gamma_t + \gamma_H\right) - \Omega\gamma_H. \tag{7}$$

Therefore, the slope and the y-intercept of a fit to a line of the frequency vs. γ_H should allow us to determine Ω.

4 Questions

Marc had a whole list of questions regarding this scheme. I'll attempt to reconstruct it and add a few of my own along the way.

1. From eq. (2), Ω depends upon the population distribution of the hydrogen atoms in the maser. When we fit the maser frequency shift ($\Delta\omega$) vs. the density dependent linewidth (γ_H), the parametric variable is the beam flux and thus the density. This could potentially change the state distribution in the maser and thus Ω. While the UBC experiment worried about these issues they found that their plots looked linear [3]. Will this be an issue for us?

2. What effect do magnetic gradients have on this experiment? Both Ron [1] and the UBC folk [3] (esp. pages 2504-2505) discuss the "magnetic inhomogeneity shift" or MIS. This shift can apparently lead to density dependent frequency shifts which can mimic spin-exchange effects [5]. (*Is it possible that it is merely a breakdown in the assumption that $\gamma_2' = \gamma_1'$? I'll have to read more of Crampton's article.*) The UBC folk found that their gradients were small enough that it wasn't a problem.

3. How do we determine η, the filling factor, and V_c the cavity volume, given that we would rather not open the copper pot again? In principle, we know the volume of bulb and the volume of the sapphire so perhaps we can determine these numbers with some accuracy. These parameters are necessary for determining A in eq. (5) and thus γ_t. However, given that q is small, we may not be that sensitive to these parameters.

4. How well must we be able to reset the cavity frequency to perform a reasonable measurement of Ω? (At this point, we should ask what accuracy we would like in our measurement of Ω itself. I would guess a 20% measurement would be sufficient. That's about how well Ron did at room temperature [7] and it's perhaps a bit better than the UBC folk did [3] (see page 1601).) Also, my present understanding is that the main need for resetability is to correct for maser frequency drift. Perhaps, if we can measure cavity frequencies and pulling with sufficient accuracy, we can live without resetability.

5. Marc points out that the output power of the cryomaser falls off over several hours of use. After heating the nozzle overnight the power level can be most of the way recovered. This may cause problems when we try to measure Ω as returning the flux to a preet level will not get us constant power. More troubling, we don't know whether the state distribution is affected when the power drops. (Are we getting a constant hydrogen flux but fewer atoms in the $F = 1$, $m_F = 0$ state?) If this is the case, it could also change the value of Ω which is a function of $\rho_{cc} + \rho_{aa}$.

6. Marc observed problems on his last successful cooldown in which changing the mechanical tuner heated the maser pole. He thought he could correct for this effect by locking the maser pole temperature with its heater. Will we be able to live with this? What is causing it?

5 Plug in Numbers

Let's try to plug in numbers into frequency shifts using nominal paramters as best we can determine them. Repeating equation (1)

$$2\pi(\nu - \nu_0) = \left[\Delta_c + \alpha\bar{\lambda}_{se}\right]\gamma_2 - \Omega\gamma_H \tag{8}$$

where $\Delta_c = (2Q_c/\omega_0)(\delta\omega_c)$. For our cavity, $Q \approx 4 \times 10^4$ and $\Delta_c = 6 \times 10^{-5} \cdot \delta\nu_c$.

The line-Q for our maser was measured [8] as $Q_l \approx 2.3 \times 10^9$ which implies that $\gamma_2 \approx 1.9\ \mathrm{s}^{-1}$. (With the quartz bulb in place, Marc has measured $Q_l \approx 6 \cdot 10^9$ which reduces the full linewidth to below $1\ \mathrm{s}^{-1}$.) As we haven't measured a value for γ_t, yet, for the cryomaser, we don't know how to separate γ_H from the other decoherence processes. However, if we look at numbers for a room temperature maser [4], we find that $\gamma_2(RT) \approx 1.5\ \mathrm{s}^{-1}$ while $\gamma_t(RT) \approx 1.35\ \mathrm{s}^{-1}$. This leaves only $0.15\ \mathrm{s}^{-1}$ of possible rate for density dependent processes. Of course in the cryomaser, we would expect it to be smaller, still.

We can also attempt to estimate our expected value of γ_H directly from values determined by Hardy. Following Verhaar, we know that

$$\gamma_H = [\sigma_1(\rho_{aa} + \rho_{cc}) + \sigma_2] \cdot \bar{v} n_H \tag{9}$$

where the σ are Verhaar coefficients, $\bar{v} = \sqrt{16kT/\pi m} \approx 14,000$ cm/s is the mean thermal velocity and $n_H \approx 10^9\ \mathrm{cm}^{-3}$ is the hydrogen density in the maser bulb [8]. Hardy determines that $\sigma_1(\rho_{aa} + \rho_{cc}) + \sigma_2 \approx 0.4$ Å^2 or $4 \times 10^{-17}\ \mathrm{cm}^2$. Therefore, we expect that $\gamma_H \approx 6 \times 10^{-4}\ \mathrm{s}^{-1}$ which is substantially smaller than the estimate at room temperature above.

Since Verhaar and co-workers calculate $\Omega \approx 0.08$ for state selection which produces $\rho_{aa} + \rho_{cc} = 0.5$ which we should approach at low densities and properly working beam state-selection. Hardy and company at UBC measured a value of $\Omega \approx -0.06$ which, while disagreeing in sign, is of the same magnitude as the theoretical value. We can, therefore, use either of these as an estimate of the required sensitivity.

Finally we would like to have the standard DIS values for the cryomaser. These may be more difficult to estimate as we need various parameters that haven't been measured for the maser. I had some problems finding an equation into which to plug numbers. I fear that various Hardy, Verhaar and Crampton equations for the DIS h-i shift have errors in them. Since we only have the temperature dependence of the cross section from Verhaar [9] we should begin there. In eq. (29-32), they write that the standard DIS shift is

$$\Delta\omega_{\mathrm{DIS}} = \left[\gamma\bar{v}\bar{\lambda}_0(1 + \Delta_c^2)\right]\gamma_2 \tag{10}$$

where ω_{DIS} is the DIS frequency shift, $\bar{v}$ is the mean thermal velocity, Δ_c is the cavity detuning in the usual units, $\bar{\lambda}_0$ is Verhaar's cross section, and γ is the mystery parameter, not defined by Verhaar.

Happily, Marc worked it out from Vanier and Audoin [10]. We begin by repeating Verhaar's equation (29) which more or less defines γ:

$$\frac{N}{V_b}(\rho_{cc} - \rho_{aa}) = \gamma\left(1 + \Delta^2\right)\Gamma \tag{11}$$

where N is the number of atoms in the bulb, V_b is the bulb volume (thus their ratio is the hydrogen density) and Γ is the total linewidth. This relation is clearly related to the self-consistency of the cavity field and the field generated by the atoms. The difficulty with this relationship, as stated above, is that we don't know γ. However, Vanier and Audoin come to our rescue. In the first chapter on hydrogen masers, we find eq. (6.3.3) (rewritten in my notation):

$$\rho_{cc} - \rho_{aa} = \frac{1}{N} \frac{T_1 I \left[1 + T_2^2(\omega - \omega_0)^2\right]}{1 + T_1 T_2 b_s^2 + T_2^2(\omega - \omega_0)^2} \tag{12}$$

which clearly comes from the magnetic field self-consistency. In this equation, I is the total atomic flux into the bulb and b_s is going to drop out in the next step. We can substitute into this equation, eq. (6.3.10) which equates then denominator of the above equation to $KQ_cT_1T_2I$. A bunch of constants have been absorbed into $K = \frac{\mu_0 \mu_B^2 \eta}{\hbar V_c}$ where $\mu_0 = 4\pi \times 10^{-7}$ N/A^2 is the permitivity of space, $\mu_B = 9.3 \times 10^{-24}$ J/T is the Bohr magneton, η is the filling factor, and V_c is the cavity volume. Inserting eq. (6.3.10) into eq. (12) we find

$$\rho_{cc} - \rho_{aa} = \frac{1}{N} \frac{T_1 I \left[1 + T_2^2(\omega - \omega_0)^2\right]}{KQ_cT_1T_2I}. \tag{13}$$

We can rewrite $T_2(\omega - \omega_0) = 2Q_l/\omega_0\ (\omega - \omega_0)$ and then use the line pulling equation in the form of $Q_l\Delta\omega = Q_c\Delta\omega_c$ and further simplify to find that

$$\rho_{cc} - \rho_{aa} = \frac{1}{N} \frac{\left[1 + \Delta_c^2\right]}{KQ_cT_2} \tag{14}$$

where $\Delta_c = 2Q_c/\omega_0\ \Delta\omega_c$ which Verhaar simply calls Δ. We can now compare eq. (14) with eq. (11) above from Verhaar to determine that

$$\gamma = \frac{1}{V_bKQ_c} = \frac{\hbar V_c}{\mu_0 \mu_B^2 \eta V_b Q_c} \tag{15}$$

where all the parameters were defined above. We can further simplify this expression by remembering that $\eta' = \eta V_b/V_c$ and thus

$$\gamma = \frac{\hbar}{\mu_0 \mu_B^2 \eta' Q_c} \tag{16}$$

where η' is the modified filling factor defined [11] as

$$\eta' = \frac{\langle H_z \rangle_{\text{bulb}}^2}{\langle H_z^2 \rangle_{\text{cavity}}} \frac{V_b}{V_c} \tag{17}$$

where H_z is the amplitude of the microwave field which will be averaged over the appropriate volume. In eq. (28) of the same paper, they point out that for cylindrical bulb of radius r and length L and a cylindrical cavity of corresponding dimensions R and L that

$$\eta' = \frac{32}{\pi} \frac{J_1^2(kr)}{J_0^2(kR)} \frac{sin^2(\pi/2)(l/L)}{1 + (\pi/kL)^2} \frac{L}{lk^2R^2} \tag{18}$$

which should be handy in determining the cryomaser filling factor.

We now have almost enough information to determine the DIS frequency shift. Rewriting eq. (10) with our new expression for γ, we find that

$$\Delta\omega_{\text{DIS}} = \left[\frac{\hbar}{\mu_0 \mu_B^2 \eta' Q_c} \bar{v} \bar{\lambda}_0 (1 + \Delta_c^2)\right] \gamma_2. \tag{19}$$

We still need an estimate for the filling factor, η', but all the other parameters are known.

Plugging numbers into eq. (19) in MKS units, we find that

$$\alpha\bar{\lambda}_{se} = \gamma\bar{v}\bar{\lambda}_0 \quad (20)$$

$$= \frac{\hbar\bar{v}\bar{\lambda}_0}{\mu_0\mu_B^2\eta' Q_c} \quad (21)$$

$$= \frac{1\cdot 10^{-34}\ (-1.7\cdot 10^{-17}\mathrm{m}^3/\mathrm{s})}{4\pi\cdot 10^{-7}\ (9.3\cdot 10^{-24})^2\ \eta'\ 40000} \quad (22)$$

$$= 4\cdot 10^{-4}/\eta' \quad (23)$$

leaving η', which is probably between 5 and 10, left to be determined. This gives us $\alpha\bar{\lambda}_{se} \approx 10^{-4}$ for the cryomaser.

Now, let's go back to eq. (1) and actually put the numbers in.

$$2\pi\Delta\nu_{\mathrm{mas}} = \left[\frac{2Q_c}{\omega_0}\Delta\omega_{\mathrm{cav}} + \alpha\bar{\lambda}_{\mathrm{se}}\right]\gamma_2 - \Omega\ \gamma_H \quad (24)$$

$$= \left[5.7\cdot 10^{-5}\Delta\nu_{\mathrm{cav}} + \frac{4\cdot 10^{-4}}{\eta'}\right]\left(1\ s^{-1}\right) \quad (25)$$

$$-(0.07)\ (6\cdot 10^{-4}\ \frac{n_H}{10^9}) \quad (26)$$

Therefore, we're expecting that as we vary the maser flux and change the density from the maximum observed to date to zero, we expect a change in the maser frequency, $\Delta\nu_{\mathrm{mas}} = 6.7\cdot 10^{-6}$ Hz. Therefore we need a frequency stability of nearly $\Delta\nu/\nu \approx 10^{-15}$ to resolve these shifts. Of course increasing the flux and thus hydrogen density by an order of magnitude would make these shifts much easier to observe.

We can also notice that the DIS shift $\alpha\bar{\lambda}_{\mathrm{se}} \approx 1\cdot 10^{-5}$ Hz which we should compare to the room temperature maser. The nominal DIS tuning of the maser cavity would only correxpond to about a 10 Hz detuning. Therefore, it will be very hard to extract this value from our data.

References

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