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Hydrogen Spin Exchange Frequency Shifts

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Helmut Hellwig

Frequency shifts due to hydrogen spin exchange collisions are discussed for the hydrogen maser and the hydrogen storage beam tube. The combined effects of spin exchange and cavity pulling in the hydrogen maser are evaluated with emphasis on frequency errors introduced by standard tuning procedures. It is found that an automatic cavity tuning system based on a variation of the linewidth does not introduce a frequency error. In contrast, manual tuning procedures based on an interpolation of measurements at different cavity settings do not yield a perfect compensation of spin exchange effects. However, the frequency error becomes only significant at unnecessarily large cavity offsets.

The smallness of the spin exchange shifts and the near absence of cavity pulling in a hydrogen storage beam tube are expected to permit a determination of spin exchange shifts by varying the beam intensity with a precision adequate for stability and accuracy figures of 10^{-14} or better.

Key Words: Frequency pulling; hydrogen beam tube; hydrogen maser; maser tuning; spin exchange.

1. INTRODUCTION

Collisions between radiating hydrogen atoms cause a broadening of the resonance line as well as a frequency shift. An important feature of the hydrogen maser is the automatic cancellation of the spin exchange frequency shift by cavity pulling if the hydrogen maser tuning method is based on varying the hydrogen relaxation time. The most widely used method is the beam intensity modulation technique (Vanier and Vessot, 1964).

A need was felt to discuss and recheck this result. An additional impetus for reevaluating this aspect of the hydrogen maser was provided by the fact that only one short discussion of spin exchange and cavity pulling is published in the open literature (Crampton, 1967) although the result is widely used in experiments with the hydrogen maser (Vanier and Vessot, 1964).[1] Furthermore, a quantitative estimate of the spin exchange frequency shift to be expected in the proposed hydrogen storage beam tube was needed (Hellwig, 1970).

The following discussion starts from basic equations for spin exchange effects given by Bender (1963). The treatment differs somewhat from the one given by Crampton (1967); in particular a detailed discussion of the cavity pulling in combination with spin exchange shifts is given, and the frequency errors associated with maser tuning techniques are evaluated.

2. SPIN EXCHANGE FREQUENCY SHIFT

The shifted frequency[2] ω' differs from the unperturbed transition frequency ω_0 by an amount proportional to the rate of spin exchange phase shifting V and to the population difference $(\rho_{22} - \rho_{44})$ of the two $m_F = 0$ levels. The relationships are given by (19), (22), and (24) in Bender's article (1963)

$$\omega' - \omega_0 = -\tfrac{1}{4} V (\rho_{22} - \rho_{44}) \tag{1}$$

$$V = 2\pi n \bar{v}_{rel} \int_0^\infty \{\sin \Delta (R)\} R \, dR \tag{2}$$

[1] Every publication giving a frequency determination of the hydrogen maser relies on the automatic cancellation of the spin exchange shift by cavity pulling when the maser is tuned.

[2] Throughout this paper ω is just called "frequency" although, in a strict sense, it is the angular frequency.

$$\rho_{22} - \rho_{44} = \frac{K^2 + (\omega - \omega')^2}{[K^2 + (\omega - \omega')^2][2 + U/r] + \frac{8\beta^2 K}{r}} . \tag{3}$$

The symbols have the following meaning: ω = running frequency; n = atomic density; $\bar{v}_{rel}$ = relative atomic velocity; R = collision parameter; $\Delta(R)$ = phase shift; K = total relaxation constant; U = rate of spin exchange collisions; β = rf magnetic field parameter; r = rate of escape of atoms from the bulb. K and β can be expressed as

$$K = r + \tfrac{1}{4} U \tag{4}$$

$$\beta = \frac{\mu_0 \langle H_z \rangle_b}{2\hbar} \tag{5}$$

where μ_0 = magnetic dipole moment of the transition, and $\langle H_z \rangle_b$ = average magnetic rf field in the bulb region. It must be noted that relaxation processes other than escape from the bulb and spin exchange are not included in this discussion.

3. CONDITIONS FOR THE HYDROGEN STORAGE BEAM TUBE

The hydrogen storage beam tube (Hellwig, 1970) is a proposed new device which potentially combines the advantages of the hydrogen maser with those of a beam tube. Its concept is a logical extension of Ramsey's original storage beam proposal and his "bounce box" experiments with cesium (Kleppner, Ramsey, and Fjelstad, 1956) which led to the development of the hydrogen maser (Kleppner, Goldenberg, and Ramsey, 1962).

The hydrogen storage beam tube resembles essentially the configuration of a hydrogen maser, however it is operated well below oscillation threshold, and an external microwave signal is injected into the cavity. The number of atoms leaving the bulb which have undergone a transition is monitored with a second selector magnet and a beam detector in a fashion quite similar to the standard operational principle of an atomic beam tube.

Of the various aspects of the hydrogen storage beam tube the one which is of importance to the discussion presented here is the near absence of cavity pulling. For passive beam tube devices one obtains approximately a pulling which is proportional to the ratio of the linewidths of cavity and atomic transition <u>squared</u> (Holloway and Lacey, 1964) whereas the pulling in a maser is directly proportional to this ratio as can be seen from (20) below. As a result the cavity pulling in the hydrogen storage beam tube can be expected to be reduced by many orders of magnitude as compared to the hydrogen maser (Hellwig, 1970).

In the following we shall estimate the spin exchange frequency shift to be expected in a hydrogen storage beam tube.

We assume that beam intensities of 10^{12} atoms per second at storage times of about 1 s shall not be exceeded.[3] For these conditions the spin exchange line broadening is small compared to the linewidth due to the storage time. We therefore may use the approximations

$$U/r << 1 \text{ and } K \approx r \quad .$$

[3] For a bulb of 10^3 cm^3 volume this corresponds to an atomic density of about 10^9 atoms per cm^3.

We furthermore restrict ourselves to microwave driving frequencies well within the linewidth $\delta\omega' = 2K$ of the transition. We may then use the approximation

$$\omega - \omega' << K.$$

With the above approximations (3) simplifies to

$$\rho_{22} - \rho_{44} \approx \frac{r^2}{2r^2 + 8\beta^2} = \frac{1}{2+8(\beta/r)^2} \quad . \tag{6}$$

The power of the exciting microwave signal will approximately be chosen as

$$\frac{\beta}{r} = \frac{\pi}{2} \quad . \tag{7}$$

Bender (1963) evaluated numerically the integral in (2), and gave as a result

$$V \approx 13\pi a_0^2 n \bar{v}_{rel} \tag{8}$$

where a_0 is the Bohr radius ($a_0 = 0.529 \times 10^{-8}$ cm). After combining (1), (6), (7), and (8) we obtain with $\bar{v}_{rel} = 2 \times 10^5$ cm/s

$$\omega' - \omega_0 = kn \tag{9}$$

where $k \approx -2.6 \times 10^{-12}$ cm^3/s, and n is measured in cm^{-3}.

The spin exchange frequency shift is proportional to the atomic density n, and its value for the assumed highest density of $n = 10^9$ atoms/cm^3 is $\omega' - \omega_0 \approx -2.6 \times 10^{-3}\ s^{-1}$ or a fractional frequency shift of about 3×10^{-13}. According to (9) this frequency shift is linear with the atomic density and can therefore be measured conveniently by varying the beam intensity. It has to be noted that a variation of the beam intensity causes only negligible cavity pulling due to the near-absence of this effect in the beam tube device as was pointed out previously.

4. CONDITIONS FOR THE HYDROGEN MASER OSCILLATOR

The condition for self-sustained maser oscillations can be written as

$$P_{rad} = P_{abs} \tag{10}$$

where the radiated power is given by

$$P_{rad} = n V_b \hbar \omega \frac{d\rho_{22}}{dt} \tag{11}$$

and the absorbed power by

$$P_{abs} = \frac{\omega}{Q} \frac{V_c}{8\pi} \langle H^2 \rangle_c \ . \tag{12}$$

V_b is the bulb volume, V_c the cavity volume, $d\rho_{22}/dt$ the rate of emission from one atom, and $\langle H^2 \rangle$ the average squared rf magnetic field in the cavity. The rate of emission was given by Bender (1963) as

$$\frac{d\rho_{22}}{dt} = \frac{2\beta^2 K}{[K^2 + (\omega - \omega')^2][2 + U/r] + \frac{8\beta^2 K}{r}} . \qquad (13)$$

The combination of (10) to (13) yields

$$[K^2 + (\omega - \omega')^2][2 + U/r] + \frac{8\beta^2 K}{r} = \frac{16\pi \hbar Q V_b \beta^2}{V_c \langle H^2 \rangle_c} nK . \qquad (14)$$

β^2 in (14) can be substituted from (5). The filling factor η' as defined by Kleppner and Vessot et al. (1965) is given by

$$\eta' = \frac{\langle H_z \rangle_b^2 V_b}{\langle H^2 \rangle_c V_c} . \qquad (15)$$

We introduce the parameter λ' which contains only constants for a given maser configuration

$$\lambda' = \frac{\hbar}{16\pi \mu_0^2 \eta' Q} . \qquad (16)$$

Equations (14) to (16) combine with (1), (3), and (5) to

$$\omega' - \omega_0 = -\lambda' \frac{V}{n} [1 + (\frac{\omega - \omega'}{K})^2] K . \qquad (17)$$

Inserting (8) into (17) leads to

$$\omega' - \omega_0 = -13\pi a_0^2 \bar{v}_{rel} \lambda' [1 + (\frac{\omega - \omega'}{K})^2] K . \qquad (18)$$

If we restrict the frequencies to a small range around ω' we obtain with $C = 13\pi a_0^2 \bar{v}_{rel} \lambda'$

$$\omega' - \omega_0 = -CK . \qquad (19)$$

Equations (17) and (18) do not contain explicitly the beam intensity; instead, the spin exchange frequency shift is proportional to the total linewidth $\delta\omega_a = 2K$. At first the result (19) is surprising because one would expect $(\omega' - \omega_0) \to 0$ for $\delta\omega_a \to \delta\omega_E$ where $\delta\omega_E$ is the line-width due to escape from the bulb alone (vanishing contribution of spin exchange effects). However, (18) does not permit the limit of vanishing spin exchange effects. The equation was derived using the condition (10) for self-sustained maser oscillations and is therefore only valid under these conditions. In order to realize an oscillating maser in the limit $\delta\omega_a \to \delta\omega_E$, the parameter[4] λ' has to approach zero,and $(\omega' - \omega_0) \to 0$ according to (18).

[4] λ' relates to the maser oscillation parameter q, which was defined by Kleppner and Vessot et al. (1965).

5. TUNING OF THE HYDROGEN MASER

The pulling of the maser output frequency by a cavity which is detuned from the atomic transition frequency is one of the earliest discoveries in maser theory (Gordon, Zeiger, and Townes, 1955), and is quantitatively given by (Shirley, 1968)

$$\omega = \frac{\delta\omega_c \omega_a + \delta\omega_a \omega_c}{\delta\omega_c + \delta\omega_a} \qquad (20)$$

where $\delta\omega_c$ and $\delta\omega_a$ are the linewidths of cavity and transition line, respectively, and ω_a is the atomic transition frequency. Spin exchange is included by substituting ω' for ω_a. We also substitute (Q = loaded cavity Q)

$$\delta\omega_c = \frac{\omega_c}{Q} \text{ and } \delta\omega_a = 2K$$

and obtain

$$\omega - \omega' = \frac{K}{(\omega_c/2Q)+K} (\omega_c - \omega') . \qquad (21)$$

ω' can be expressed from (18), and (21) then reads after some simple regrouping

$$\omega - \omega_0 = \frac{K}{(\omega_c/2Q)+K} \left[\omega_c - \omega_0 - \frac{\omega_c}{2Q} C \left(\frac{\omega - \omega'}{K}\right)^2\right] . \qquad (22)$$

If we use the approximations

$$\frac{\omega_c}{2Q} >> K \text{ and } \omega - \omega' << K$$

(22) simplifies to

$$\omega - \omega_0 = \frac{2Q}{\omega_c} K(\omega_c - \omega_0 - \frac{\omega_c}{2Q} C) . \tag{23}$$

Equation (23) is the standard "tuning equation" for the hydrogen maser (Vanier and Vessot, 1964). If the maser is tuned so that a variation of K does not cause a change in the maser output frequency then the maser frequency is equal to the unperturbed atomic transition frequency ($\omega = \omega_0$), and the cavity is detuned by an amount equal to the spin exchange frequency shift ($\omega_c - \omega_0 = C\,\omega_c/2Q$) as can be seen from (23).

The error committed in tuning the maser in this fashion can be evaluated from the above approximations. Omitting K in the denominator of the "pulling factor" in (22) does not introduce any error in the tuning procedure. The omission of the last, K-dependent, term in (22) seems to introduce a frequency error. The K-dependent part of this term can be rewritten using (19)

$$\left(\frac{\omega - \omega'}{K}\right)^2 = \left(\frac{\omega - \omega_0 + CK}{K}\right)^2 = \left(\frac{\omega - \omega_0}{K} + C\right)^2 . \tag{24}$$

The constant C in this expression does not introduce an error in the above tuning method. The K-dependent part $(\omega - \omega_0)/K$ approaches zero as the maser is tuned ($\omega \to \omega_0$). As a result no appreciable frequency error is introduced by a tuning procedure which uses the variation of K.

This is only true for experiments where the maser is actually set to oscillate on the transition frequency ω_0 as for example in automatic servo tuning systems (Hellwig and Pannaci, 1967; and Vessot, Levine, Mueller,and Baker, 1967). Frequently, however, the maser frequency is only indirectly determined using two or more cavity settings at which K is varied (Vanier and Vessot, 1964). The maser frequency is then calculated using a linear interpolation between the measured variations of the maser output frequency. In this case a frequency error is introduced which corresponds to the variation with K of the last term in (22). This term can be written as $-C \frac{(\omega - \omega')^2}{K}$. The error follows then approximately as $C \frac{(\omega - \omega')^2}{K^2} \Delta K$, where ΔK is the variation of K. The maser constant C has typically a value of $C \approx 10^{-2}$. Fortunately, the error has an appreciable magnitude only if fairly large cavity offsets and large K-variations are used. For example a cavity setting corresponding to a fractional maser frequency offset of 10^{-10} will produce a fractional error of about 10^{-13} if $\Delta K \approx K \approx 1\ s^{-1}$.

6. CONCLUSIONS

The discussion given in this report indicates that an automatic cavity servo control based on varying the linewidth of the atomic transition yields an exact tuning of the maser. The spin exchange frequency shift will be perfectly compensated by a proper cavity detuning. A manual tuning procedure in which the maser tuning is found by interpolating between measurements at different cavity settings will introduce in general a systematic error due to an incomplete compensation of frequency shifts. However, this error only has an appreciable magnitude if unnecessarily large cavity offsets are used.

It was pointed out that the automatic compensation of spin exchange and cavity pulling effects, as described by (23), is only valid for an oscillating maser.

In a passive device such as the proposed hydrogen storage beam tube the frequency shift due to spin exchange is present. The shift is linear with the beam intensity (atomic density in the storage bulb) and can be measured and corrected for by varying the beam flux. The magnitude of the shift is small; of the order of 10^{-13} for typical beam intensities and storage times. Thus only a moderate measurement precision and a moderate control of the beam flux is necessary if stabilities and accuracies of 10^{-14} are the design goal of a passive device (Hellwig, 1970).

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