Measurement of the Hydrogen Spin-Exchange Cross Section with the Hydrogen Maser

H. HELLWIG
Electronic Components Laboratory, United States Army Electronics Command, Fort Monmouth, New Jersey
(Received 25 August 1967)

The oscillation characteristics of the hydrogen maser under the influence of hydrogen-hydrogen spin-exchange collisions are described and discussed. Using experimental data from an oscillating hydrogen maser, the hydrogen spin-exchange cross section is calculated. A value of $\sigma=2.65\times10^{-15}$ cm^2 at room temperature is obtained and compared with previously published values. A brief discussion on a possible increase of the maser output power is given.

INTRODUCTION

THE hydrogen maser has not only been applied very successfully as a frequency standard[1,2] but as well as a precision tool for the measurement of physical quantities.[2,3]

This paper describes the theory and experiments leading to the determination of the hydrogen spin-exchange cross section.

The basic theory, techniques and design principles relevant to the hydrogen maser have been published in two papers, one by Kleppner, Goldenberg and Ramsey,[4] the other being a joint effort of two groups at Harvard University and Varian Associates.[5] This paper is based on these two publications hereafter referred to as KGR[4] and HV[5], respectively.

The performance of the hydrogen maser, especially its behavior with changing hydrogen beam intensity, is largely influenced by spin-exchange collisions between the hydrogen atoms in the storage bulb. In the following second section the formulas describing the operating conditions of the hydrogen maser are derived. The third section deals with the determinations of the hydrogen spin-exchange cross section and its comparison with previously published values.

MASER OSCILLATIONS AND SPIN EXCHANGE

The first task is to include the effect of hydrogen spin exchange in the equations which describe the behavior of the hydrogen maser, in particular into the equation for the threshold beam intensity. The minimum beam intensity required for the threshold of oscillation is given by Eq. (14) in KGR.[4]

$$I_{\mathrm{th}}=c_1\gamma^2\,, \tag{1}$$

with $c_1=hV_c/8\pi^2\mu_0{}^2Q\eta$, where V_c is the volume, Q the loaded-cavity Q factor, and η the filling factor[4] of the cavity. γ is the over-all relaxation constant and includes the spin-exchange relaxation, thus making I_{th} dependent on this process, that is, dependent on the actual beam intensity.

Regarding the fact that one has to distinguish carefully between relaxation of the population difference between the two maser levels described by γ_1, and relaxation of the oscillating moment described by γ_2, one arrives at

$$\gamma^2=\gamma_1\gamma_2\,, \tag{2}$$

where γ_1 and γ_2 are each the sum of the relaxation constants of all corresponding relaxationpr ocesses. For the hydrogen spin-exchange (SE) process it can be shown that $\gamma_{1\mathrm{SE}}=2\gamma_{2\mathrm{SE}}$. Assuming that all other relaxation processes including the escape from the bulb can be described by $\gamma_{1t}=\gamma_{2t}=\gamma_t$ one gets from Eq. (2)

$$\gamma^2=\tfrac{1}{2}\gamma_{1\mathrm{SE}}{}^2+\tfrac{3}{2}\gamma_{1\mathrm{SE}}\gamma_t+\gamma_t{}^2\,. \tag{3}$$

The spin-exchange relaxation constant can be expressed by the spin-exchange cross section σ, the relative average velocity $\bar{v}_r$ and the particle density in the bulb ρ:

$$\gamma_{1\mathrm{SE}}=\rho\bar{v}_r\sigma\,. \tag{4}$$

Substituting $\rho=I_{\mathrm{tot}}/\gamma_bV_b$ in Eq. (4), where γ_b and V_b are the escape relaxation constant and the volume of the storage bulb, respectively, and I_{tot} is the total beam intensity one gets

$$\gamma_{1\mathrm{SE}}=\frac{c_2}{\gamma_b}I\,, \tag{5}$$

where $c_2=2\sigma\bar{v}_r/V_b$. The factor 2 originates from the fact that, assuming ideal state selection and beam optics, not only the atoms in the $F=1$, $m=0$ state (beam intensity I) which is the origin of the maser transition are focused into the bulb, but also an equal amount of atoms in the $F=1$, $m=1$ state.[4]

After combining Eq. (1), (3), and (5) the dependence of the threshold beam intensity on the actual beam intensity is given by

$$I_{\mathrm{th}}=2c_1c_2{}^2\frac{1}{\gamma_b{}^2}I^2+3c_1c_2\frac{\gamma_t}{\gamma_b}I+\gamma_t{}^2\,. \tag{6}$$

The physical meaning of Eq. (6) is qualitatively depicted in Fig. 1. Plotted is the threshold inten-

[1] R. Vessot, H. Peters, J. Vanier, R. Beehler, D. Halford, R. Harrach, D. Allan, D. Glaze, C. Snider, J. Barnes, L. Cutler, and L. Bodily, Trans. IEEE **IM-15**, 165 (1966).
[2] N. F. Ramsey, Metrologia **1**, 7 (1965).
[3] N. F. Ramsey, in Proceedings of the 21st Annual Symposium on Frequency Control, Atlantic City, N. J., 1967, p. 500 (unpublished).
[4] D. Kleppner, H. M. Goldenberg, and N. F. Ramsey, Phys. Rev. **126**, 603 (1962).
[5] D. Kleppner, H. C. Berg, S. B. Crampton, N. F. Ramsey, R. F. C. Vessot, H. E. Peters, and J. Vanier, Phys. Rev. **138**, A972 (1965).

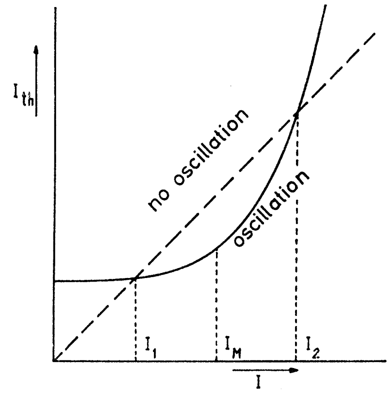

FIG. 1. Threshold of oscillation as a function of the actual beam intensity.

sity I_{th} as a function of the actual beam intensity I according to Eq. (6). The straight line $I_{th}=I$ divides the diagram in two sections. If the curve is in the section above $I_{th}=I$, no maser oscillations are possible whereas below $I_{th}=I$ oscillations will occur. The region of oscillations for the hydrogen maser can therefore be described by $I_1 \leqslant I \leqslant I_2$. For beam intensities $I<I_1$ the energy carried by the beam into the cavity is not sufficient for self-sustained oscillations, and for intensities $I>I_2$ the increase in the number of spin-exchange collisions prevents oscillations.

By setting $I_{th}=I$ in Eq. (6) one easily gets

$$I_{1,2}=\frac{\gamma_b^2}{4c_1c_2^2}\left\{1-3\frac{\gamma_t}{\gamma_b}c_1c_2 \mp\left[1-6\frac{\gamma_t}{\gamma_b}c_1c_2+\left(\frac{\gamma_t}{\gamma_b}c_1c_2\right)^2\right]^{1/2}\right\}. \quad (7)$$

Equation (7) corresponds to Eq. (17) in HV[5] but was derived here in a somewhat different way. As in HV the necessary condition which has to be fulfilled if oscillations shall be possible is [from Eq. (7)]

$$6(\gamma_t/\gamma_b)c_1c_2<1.032. \quad (8)$$

The output power of the hydrogen maser is given by the combination of Eq. (14) and (16) in KGR as

$$P=c_3(I-I_{th}) \quad (9)$$

with $c_3=\omega(h/4\pi)(Q/Q_1)$, where ω is the transition frequency and Q_1 represents the output coupling. Substituting I_{th} in Eq. (9) by Eq. (6) the beam intensity I_M which will give maximum output power can be derived. From $dP/dI=0$ it follows:

$$I_M=\tfrac{1}{2}(I_1+I_2)=\frac{\gamma_b^2}{4c_1c_2}\left(1-3\frac{\gamma_t}{\gamma_b}c_1c_2\right). \quad (10)$$

Inserting Eq. (10) in Eq. (6) one gets

$$I_{th}=\frac{\gamma_b^2}{8c_1c_2}\left[1-\left(\frac{\gamma_t}{\gamma_b}c_1c_2\right)^2\right] \quad (11)$$

and from Eqs. (9) through (11) the maximum output power attainable with a given set of maser parameters is obtained as

$$P_M=\frac{\gamma_b^2c_3}{8c_1c_2}\left[1-6\frac{\gamma_t}{\gamma_b}c_1c_2+\left(\frac{\gamma_t}{\gamma_b}c_1c_2\right)^2\right]. \quad (12)$$

Equation (12) shall be rewritten inserting the expressions for c_1 through c_3 and using the following two approximations: (a) The bulb escape relaxation shall be the dominating process, that is $\gamma_t \approx \gamma_b$ with $\gamma_b=\bar{v}_rA/4\sqrt{2}V_b$, where A is the effective escape area of the bulb. (b) The quadratic term in the brackets of Eq. (12) shall be neglected. It follows:

$$P_M=\frac{\pi\omega\mu_0^2}{128\sigma^2}\frac{Q^2A^2\eta}{Q_1V_c}\left(1-\frac{h\sigma\bar{v}_r}{4\pi^2\mu_0^2}\frac{V_c}{V_b\eta Q}\right). \quad (13)$$

Equation (13) suggests a brief discussion of the output power of the hydrogen maser. The second term in the parenthesis of Eq. (13) has a value of about 0.5 to 0.6 for hydrogen masers of conventional design.[6] Assuming that higher output power is desired, optimizing this term yields an increase in output power of ultimately a factor of only 2. The term in front of the parenthesis in Eq. (13) at optimum power is essentially $Q\eta A^2/V_c$ if one disregards the constants. It is interesting to note that the above equation is independent of the volume of the bulb. The possibilities of upgrading η (smaller bulb), Q, and V_c in order to increase the power output are limited in the case of a conventional hydrogen maser, and the increase of the effective escape area A tends to degrade the total relaxation time and in connection with this the long-term stability. However, a power increase by a factor of 50 over presently used masers to about 2×10^{-11} W seems feasible, and would correspondingly increase the short-term frequency stability being still a weak point in the over-all performance of the hydrogen maser as a frequency standard. A different approach for higher output power would be the use of a cavity oscillating in a different mode (e.g., the TE_{11} mode). In this case it seems feasible to obtain the same output power as above but without degrading the relaxation time.

MEASUREMENT OF THE HYDROGEN SPIN-EXCHANGE CROSS SECTION

In order to derive the equation from which the spin-exchange cross section will be calculated, Eq. (7) shall be simplified first. The same approximation as in the

[6] For example, maser 1 in Table I of this paper.

TABLE I. Measurement data.

	Maser 1	Maser 2
r (measured)	17.4	1.20
$\bar{v}_r$ (at room temp.)	3.58×10^5 cm/sec	3.58×10^5 cm/sec
γ_b/γ_t	0.70	0.70
Q	3.40×10^4	4.0×10^4
V_b	1.94×10^3 cm^3	4.88×10^2 cm^3
V_c	1.43×10^4 cm^3	1.47×10^4 cm^3
η	2.8	6.0
σ	2.66×10^{-15} cm^2	2.64×10^{-15} cm^2

derivation of Eq. (13) is used, that is neglecting the quadratic term in the square root of Eq. (7). According to Eq. (8) this approximation will introduce a maximum error of about 3%. With $r=I_2/I_1$ being the ratio of the two critical beam intensities, the following can then be derived from Eq. (7):

$$6\frac{\gamma_t}{\gamma_b}c_1c_2=\frac{4r^{1/2}}{(1+r^{1/2})^2}. \tag{14}$$

And from this by substituting c_1 and c_2

$$\sigma=\frac{4\sqrt{r}}{(1+\sqrt{r})^2}\frac{4\pi^2\mu_0^2}{3h\bar{v}_r}\frac{\gamma_b}{\gamma_t}\frac{QV_b\eta}{V_c}. \tag{15}$$

This is the equation from which σ can be determined. It is only necessary to measure r with the maser, all other parameters are either known or can be determined quite accurately in separate measurements.

Two independent measurements were done with two different masers, one maser having a regular-size bulb, the other a small bulb of only 10-cm diameter. The ratio γ_b/γ_t was determined by calculating γ_b and measuring γ_t with microwave pulses applied to the maser operating well below threshold. The value of η was taken out of the plot of η in KGR regarding the correction pointed out in footnote 27 of HV.

TABLE II. Comparison.

Author	Origin	(10^{-5} cm^2)
Wittke and Dicke[a]	Theor. (estimate)	2.3
Bender[b]	Theoretical	2.1
Mazo[c]	Theoretical	2.85±15%
Hildebrandt, Booth, and Barth[d]	Expt. (EPR)	1.8
This paper	Expt. (H Maser)	2.65±15%

[a] J. P. Wittke and R. H. Dicke, Phys. Rev. **103**, 620 (1956).
[b] P. L. Bender, Phys. Rev. **132**, 2154 (1963).
[c] R. M. Mazo, J. Chem. Phys. **34**, 169 (1961).
[d] A. F. Hildebrandt, F. B. Booth, and C. A. Barth, J. Chem. Phys. **31**, 273 (1959).

Table I gives the necessary data for the calculation of σ from the measurements with maser 1 and maser 2. The result is

$$\sigma=2.65\times10^{-15}\text{ cm}^2\pm15\%.$$

Although the two values for σ from the two independent measurements nearly coincide, an error of 15% was estimated mainly due to uncertainty in the determination of η and γ_b/γ_t.

In the past several values for σ where published based on theoretical consideration as well as on experiments. Table II compares a selection of the most reliable data with the value reported here.

It should be noted that despite the fact that there is initially a nonequilibrium distribution of states in the hydrogen maser, the value for the spin-exchange cross section measured under this condition (this paper) agrees very well with the dependable theoretical value of Mazo calculated for an equilibrium condition.

ACKNOWLEDGMENT

The author wishes to thank E. Pannaci for his assistance in the experimental work.