

Cryogenic H maser in a strong B field

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We study the spin-exchange frequency shift of the cryogenic hydrogen maser for $B \neq 0$. A general expression is derived in terms of populations of ground-state hyperfine levels. The coefficients in this expression are calculated in the degenerate-internal-states approximation, as well as to first order in the hyperfine plus Zeeman splitting. Numerical results are compared with rigorous coupled-channel calculations. Some implications are pointed out for the frequency stability of the H maser in a magnetic field.

I. INTRODUCTION

Almost thirty years after its first realization by Goldenberg, Kleppner, and Ramsey,¹ the hydrogen maser continues to be the most stable of all frequency standards. For measuring times of about 1 h the relative frequency instability is observed to be below one part in 10^{15} . This extreme stability makes the hydrogen maser a very valuable research tool in fields as diverse as physics, astronomy, geodesy, and metrology.

As pointed out a decade ago,^{2,3} a hydrogen maser operating at liquid-helium temperature would have an even better frequency stability. This is mainly due to the much smaller collisional line broadening at lower temperatures, allowing for a larger radiating atom density, and hence for a larger radiated power without increasing the atomic linewidth above the room-temperature value. Furthermore, lower temperatures also increase the signal-to-noise ratio by decreasing thermal noise, and help to get a better control of the cavity resonance frequency. A third advantage is the possibility at sub-Kelvin temperatures to use a very reproducible wall coating of superfluid helium, with an associated wall frequency shift going through a minimum at $T=0.52$ K,⁴ which produces a very high thermal stability. Berlinsky and Hardy⁵ predicted that with such types of cryogenic hydrogen masers an improvement in frequency stability of more than two orders of magnitude over that of a room-temperature hydrogen maser should be realizable.

Up until now we did not mention the frequency shifts due to collisions between hydrogen atoms. Analysis of the effect of spin-exchange collisions⁶ showed that they shift the maser frequency in the same way as cavity pulling does: via a proportionality to the atomic linewidth. This opens the possibility to tune the cavity such that cavity pulling and spin-exchange frequency shifts compensate one another. The above-mentioned papers, however, all ignored the effect of the hyperfine energy-level separation during the collisions, which is an essential omission in the case of cryogenic H masers as was shown in two more recent papers.^{7,8} The effect of the hyperfine interaction during collisions introduces large frequency shifts which cannot be eliminated by the above spin-

exchange cavity tuning method and strongly limit the achievable stability. For a survey of the present experimental and theoretical situation we refer to Ref. 9.

In this paper we investigate the magnetic field dependence of the spin-exchange frequency shift in the H maser. This may be of interest for experiments in which the H maser is operated in a much stronger field than usual. In this context one could think in the first place of attempts to improve the frequency stability by eliminating the hyperfine-induced shift. For this application it would be essential that persistent-current solenoids and superconducting magnetic shields make it possible to operate at a much stronger constant field than usual, outside the extreme low- B regime where the first-order fluctuations of transition frequency with B vanish. It is outside the scope of the present paper to discuss the technical possibilities to keep a magnetic field of, for instance, 0.05 T stable to within a required relative variation of 10^{-18} . We confine ourselves to the question whether a magnetic field might eliminate the "dangerous" terms in the hyperfine-induced spin-exchange frequency shift. To that end it is necessary to derive the dependence of this shift on the partial densities of the four 1s hyperfine levels. At first sight it does not look improbable that such an elimination might succeed. Introducing a magnetic field of the order of 0.05 T changes the hyperfine spin wave functions and thus the collision amplitudes significantly. In addition, we will see that the symmetry lost in a collision by $B \neq 0$ introduces contributions from inelastic elements to the frequency shift besides the elastic S -matrix elements, which already play a role for $B=0$.

A second application is the present experimental activity in measuring the various contributions to the spin-exchange frequency shift. Experimental groups are interested in measuring them as a function of various experimental parameters, in particular, the atomic density in the storage bulb, to compare them with theoretical predictions but also to provide information on the population dynamics in the H maser, which is of interest for the sight into its operation. Extension of such measurements and analyses to $B \neq 0$ would enlarge the scope of present experiments. The introduction of a stronger magnetic field not only influences the collisional frequency shift, but has also a more direct influence on the maser

operation, for instance, on the maser oscillation condition.

A third type of application is associated with the use of the H maser as a precision instrument enabling one to measure very sensitively certain phenomena in atomic hydrogen gas. For some of these phenomena it may be desirable or even essential to operate the maser at stronger B fields. In this context one could think of the possibility to detect bulk or surface spin waves,¹⁰ as well as possibilities for measuring magnon effects¹¹ by means of the cryogenic H maser.

This paper is organized as follows. In Sec. II we derive the general expression for the $B \neq 0$ spin-exchange frequency shift starting from the quantum Boltzmann equation. In Sec. III we evaluate the various terms of zeroth and first order in the hyperfine level splitting by an extension of the existing method for $B=0$. In Sec. IV we present numerical results of this approach and of the rigorous coupled-channel method and discuss their application to the H maser. Some conclusions will be given in Sec. V.

II. SPIN-EXCHANGE FREQUENCY SHIFT FOR $B \neq 0$

We start from the evolution equation^{7,8} for the one-atom spin-density matrix

$$\frac{d}{dt}\rho_{\kappa\kappa'} + \frac{i}{\hbar}(\varepsilon_{\kappa} - \varepsilon_{\kappa'})\rho_{\kappa\kappa'} = \dot{\rho}_{\kappa\kappa'}|_{\text{rad}} + \dot{\rho}_{\kappa\kappa'}|_0 + \dot{\rho}_{\kappa\kappa'}|_c. \quad (1)$$

The Greek subscripts take values $a, b, c,$ and d , the ground-state hyperfine levels in order of increasing energy ε_a . The first term on the right-hand side is the radiation term resulting from the interaction of the atomic magnetic moments with the rf cavity magnetic field. The second term represents all one-atom terms such as wall collisions, finite cavity residence time, and interactions with magnetic field inhomogeneities. The third term, the collision term, is the primary point of interest in this paper. We are interested in a situation where the cavity mode is almost resonant with a particular transition $\kappa \rightarrow \kappa'$. For the corresponding density-matrix element the one-atom and collision terms then have the form

$$\dot{\rho}_{\kappa\kappa'}|_0 = -[(1/T_2)_0 - i\delta\omega_0]\rho_{\kappa\kappa'}, \quad (2)$$

$$\dot{\rho}_{\kappa\kappa'}|_c = n_H \rho_{\kappa\kappa'} \sum_{\nu} \rho_{\nu\nu} \sum_{\lambda} [(1 + \delta_{\kappa\lambda})(1 + \delta_{\kappa'\lambda})(1 + \delta_{\kappa\nu})(1 + \delta_{\kappa'\nu})]^{1/2} \langle \nu \sigma_{\kappa\kappa', \nu \rightarrow \lambda} \rangle_{\text{th}}, \quad (3)$$

in which off-resonant terms have been omitted. The complex coefficient $(1/T_2)_0 - i\delta\omega_0$ generally depends in a complicated way on the values of the diagonal density-matrix elements $\rho_{\nu\nu}$, but is independent of $\rho_{\kappa\kappa'}$. The complex "cross sections" $\sigma_{\kappa\kappa', \nu \rightarrow \lambda}$ describe the contribution of collisions in which a ν -state atom makes a transition to the λ state in colliding with an atom which is in a coherent superposition of the κ and κ' states:

$$\sigma_{\kappa\kappa', \nu \rightarrow \lambda} = \frac{\pi}{k^2} \sum_l (2l+1) \times [S_{\{\kappa\lambda\}, \{\kappa\nu\}}^l S_{\{\kappa'\lambda\}, \{\kappa'\nu\}}^{l*} - \delta_{\lambda\nu}]. \quad (4)$$

In this equation Greek subscripts between brackets are a shorthand notation for normalized symmetric (antisymmetric) two-body spin states for even (odd) partial wave l . The S -matrix elements are to be calculated for a common relative kinetic energy $E_k = \hbar^2 k^2 / m_H$ in the entrance channels $\{\kappa\nu\}$ and $\{\kappa'\nu\}$. The brackets $\langle \rangle_{\text{th}}$ in Eq. (3) denote thermal averaging.

The calculation of the collision term is based on a two-atom Hamiltonian containing, in addition to the central interaction and hyperfine interactions already included^{7,8} for $B=0$, the Zeeman term

$$V^Z = [\mu_e(\sigma_{e1} + \sigma_{e2}) - \mu_p(\sigma_{p1} + \sigma_{p2})] \cdot \mathbf{B}, \quad (5)$$

with μ_e (μ_p) the electron (proton) magnetic moment and σ the Pauli spin vector. Magnetic dipolar interactions are again negligible. The collision problem has

$\text{SO}(3)_{\text{orbit}} \times \text{SO}(2)_{\text{spin}}$ as a symmetry group, i.e., the direct product of the three-dimensional orbital rotation group and the two-dimensional spin rotation group about the z axis ($\parallel \mathbf{B}$). Due to $\text{SO}(3)$ orbital symmetry the S matrix is diagonal in the relative orbital angular momentum quantum numbers l and m_l , and independent of m_l . This is taken into account in the notation of Eq. (4). Due to $\text{SO}(2)_{\text{spin}}$ symmetry the total spin magnetic quantum number is conserved. Consequently, for odd l only elastic S -matrix elements play a role in Eq. (4), i.e., $ab \rightarrow ab$, $cb \rightarrow cb$, $ad \rightarrow ad$, and $cd \rightarrow cd$, the same combinations as for $B=0$ but with $B \neq 0$ values. For even l we have the elastic elements for $aa \rightarrow aa$, $ac \rightarrow ac$, and $cc \rightarrow cc$, and the inelastic elements for $aa \leftrightarrow ac$ and $ac \leftrightarrow cc$. The latter were absent for $B=0$ for reasons of symmetry: under a combined 180° rotation of the electron and proton spins of a single atom about an axis in the xy plane $|a\rangle \rightarrow |a\rangle$ and $|c\rangle \rightarrow -|c\rangle$, so that $|aa\rangle$ and $|cc\rangle$ are invariant and $|\{ac\}\rangle \rightarrow -|\{ac\}\rangle$. Note that by the same argument the equality of the $B=0$ elastic S -matrix elements for ab and ad and for cb and cd is lost for $B \neq 0$.

We now concentrate on the experimental situation in which $\kappa\kappa' = ac$. Substituting

$$\rho_{ac}(t) = \rho_{ac}(0) \exp[i(\varepsilon_c - \varepsilon_a)/\hbar + i\delta\omega - 1/T_2]t \quad (6)$$

in Eq. (1), we find without radiation term the total frequency shift and total atomic linewidth

$$\delta\omega = \delta\omega_0 + \delta\omega_c, \quad (7)$$

$$1/T_2 = (1/T_2)_0 + (1/T_2)_c . \quad (8)$$

Here the collisional contributions are related to the cross sections by

$$\delta\omega_c = n_H \langle v \rangle [(\rho_{cc} - \rho_{aa})\bar{\lambda}_0 + (\rho_{cc} + \rho_{aa})\bar{\lambda}_1 + \bar{\lambda}_2 + (\rho_{dd} - \rho_{bb})\bar{\lambda}_3] , \quad (9)$$

$$(1/T_2)_c = n_H \langle v \rangle [(\rho_{cc} - \rho_{aa})\bar{\sigma}_0 + (\rho_{cc} + \rho_{aa})\bar{\sigma}_1 + \bar{\sigma}_2 + (\rho_{dd} - \rho_{bb})\bar{\sigma}_3] , \quad (10)$$

with

$$\begin{aligned} i\lambda_0 - \sigma_0 &= \sigma_{ac,c \rightarrow c} - \sigma_{ac,a \rightarrow a} - \sigma_{ac,a \rightarrow c} + \sigma_{ac,c \rightarrow a} , \\ i\lambda_1 - \sigma_1 &= \sigma_{ac,c \rightarrow c} + \sigma_{ac,a \rightarrow a} + \sigma_{ac,a \rightarrow c} + \sigma_{ac,c \rightarrow a} \\ &\quad - \frac{1}{2}\sigma_{ac,b \rightarrow b} - \frac{1}{2}\sigma_{ac,d \rightarrow d} , \\ i\lambda_2 - \sigma_2 &= \frac{1}{2}\sigma_{ac,d \rightarrow d} + \frac{1}{2}\sigma_{ac,b \rightarrow b} , \\ i\lambda_3 - \sigma_3 &= \frac{1}{2}\sigma_{ac,d \rightarrow d} - \frac{1}{2}\sigma_{ac,b \rightarrow b} . \end{aligned} \quad (11)$$

Note that the new $\rho_{dd} - \rho_{bb}$ terms arise because of the above-mentioned loss of symmetry.

III. COLLISIONAL SHIFT AND BROADENING

Apparently, the collisional frequency shift and line broadening can be determined once the S matrix has been calculated. As for $B=0$, this has been done both by the coupled-channel method and by approximate methods. We refer to Refs. 7 and 8 for a description of the coupled-channel method as applied to the H maser.

A. Degenerate internal states

The calculation is much easier when energy differences of internal states are neglected. The advantage of this approximation is that the internal atomic Hamiltonian reduces effectively to a constant times the unit operator. Using this, one can turn to a new basis of internal states to simplify the collision problem, i.e., the internal basis with total electron and proton spin quantum numbers $SM_S IM_I$, which diagonalizes the interatomic interaction. With respect to this basis the coupled-channel problem reduces to a simple potential-scattering problem for singlet and triplet scattering separately.

For $B=0$ this approximation was relatively straightforward to apply. In Refs. 7 and 8 we obtained results in agreement with expressions obtained previously.⁶ It is less trivial how the approximation is to be applied most effectively to the inelastic processes for $B \neq 0$. We have shown previously^{12,13} that spin-exchange and dipolar transitions in H+H scattering in the sub-Kelvin regime can be described very successfully if one does not replace an S -matrix element as a whole by its value for degenerate internal states (DIS), but rather a related quantity: one first splits off two factors depending on initial and final channel wave numbers and subsequently approximates the remaining quantity by its value for degenerate internal states, i.e., equal wave numbers in all channels. Simple expressions in terms of scattering lengths, but still rather accurate for low energies, are obtained by calculat-

ing the remaining quantity in the zero-energy limit (vanishing wave numbers). For somewhat higher energies accurate agreement with coupled-channel values is obtained by taking the collision energy equal to the average of initial and final relative kinetic energies. Recently we applied the same so-called DIS method to the scattering of dressed H atoms in a microwave trap¹⁴ and to the reflection of H atoms from a superfluid ⁴He surface.¹⁵

As an example we give the S -matrix element for the $ac \rightarrow aa$ transition (even l),

$$S_{aa, \{ac\}}^l = (k_{aa} k_{ac} / \bar{k}^2)^{l+1/2} \sqrt{2} \times \sin 4\theta (\exp 2i\delta_T^l - \exp 2i\delta_S^l) . \quad (12)$$

In this expression k_{ac} (k_{aa}) is the initial (final) wave number, \bar{k} is their "average," δ_T (δ_S) is the triplet (singlet) scattering phase, and θ is the usual B -dependent angle characterizing the hyperfine states

$$\theta = \frac{1}{2} \arctan B_0 / B, \quad B_0 = \frac{1}{2} a / (\mu_e + \mu_p), \quad 0 < \theta \leq \frac{\pi}{4} \quad (13)$$

Making use of such expressions the λ_i and σ_i cross sections are easily obtained,

$$\begin{aligned} \lambda_0^{(\text{DIS})} &= \frac{\pi}{2k^2} (1 - 3 \cos^2 2\theta) \sum_{l \text{ even}} (2l+1) \sin 2\Delta\delta^l , \\ \lambda_1^{(\text{DIS})} &= \lambda_2^{(\text{DIS})} = 0 , \\ \lambda_3^{(\text{DIS})} &= -\frac{\pi}{k^2} \cos 2\theta \sum_{l \text{ odd}} (2l+1) \sin 2\Delta\delta^l , \\ \sigma_0^{(\text{DIS})} &= \frac{\pi}{8k^2} \sin^2 4\theta \sum_{l \text{ even}} (2l+1) \chi_-^l , \\ \sigma_1^{(\text{DIS})} &= \frac{\pi}{k^2} \sum_{l \text{ even}} (2l+1) [(1 + \cos^2 2\theta) \sin^2 \Delta\delta^l + \frac{1}{3} \sin^2 4\theta \chi_+^l] \\ &\quad - \frac{\pi}{k^2} \sum_{l \text{ odd}} (2l+1) (1 + \cos^2 2\theta) \sin^2 \Delta\delta^l , \\ \sigma_2^{(\text{DIS})} &= \frac{\pi}{k^2} \sum_{l \text{ odd}} (2l+1) (1 + \cos^2 2\theta) \sin^2 \Delta\delta^l , \\ \sigma_3^{(\text{DIS})} &= 0 , \end{aligned} \quad (14)$$

in which $\Delta\delta^l$ stands for $\delta_T^l - \delta_S^l$ and the χ coefficients for

$$\begin{aligned} \chi_+^l(k) &= (kk_> / \bar{k}_>^2)^{2l+1} \sin^2 \Delta\delta_>^l \\ &\quad + (kk_< / \bar{k}_<^2)^{2l+1} \sin^2 \Delta\delta_<^l - 2 \sin^2 \Delta\delta^l , \\ \chi_-^l(k) &= (kk_> / \bar{k}_>^2)^{2l+1} \sin^2 \Delta\delta_>^l \\ &\quad - (kk_< / \bar{k}_<^2)^{2l+1} \sin^2 \Delta\delta_<^l . \end{aligned} \quad (15)$$

In these expressions k is the wave number in the entrance channel, $k_>$ is a larger wave number obtained by adding the a - c level splitting at the magnetic field strength considered to the initial relative kinetic energy, and $k_<$ is a similar wave number found by subtracting the same splitting. Furthermore, $\bar{k}_>$ and $\bar{k}_<$ are wave numbers corresponding to averaged initial and final kinetic energies. Finally, $\Delta\delta_>^l$ and $\Delta\delta_<^l$ are the $\Delta\delta^l$ phase differences at the average wave numbers $\bar{k}_>$ and $\bar{k}_<$.

Apparently, $\lambda_1^{(\text{DIS})}$ and $\lambda_2^{(\text{DIS})}$ vanish as for $B=0$. This

is a central conclusion of the present paper. For $B=0$ we obtained dangerous λ_1 and λ_2 terms only as a correction of first order in the hyperfine level splitting. Nonvanishing values of $\lambda_1^{(\text{DIS})}$ and $\lambda_2^{(\text{DIS})}$ for $B \neq 0$ would have implied the possibility of a drastic change of these parameters already upon application of a weak magnetic field and hence, in principle, the possibility of a vanishing or density independent frequency stability parameter⁷ Ω . On the basis of the above-mentioned result we can only expect a nonvanishing value of λ_1 and λ_2 for $B \neq 0$ in first order in the hyperfine level splitting. Intuitively, one thus expects that the purpose of eliminating the effect of the dangerous terms can only be achieved with stronger B fields at least of order $B_0 \approx 50.7$ mT.

One can understand the vanishing values of $\lambda_1^{(\text{DIS})}$ and $\lambda_2^{(\text{DIS})}$ on the basis of a symmetry argument. Since the DIS two-body Hamiltonian no longer contains the two proton spins, one can carry out the above 180° rotations for electron and protons about perpendicular axes in the xy plane. This induces the transformations $|a\rangle \rightarrow |c\rangle$, $|c\rangle \rightarrow -|a\rangle$, $|b\rangle \rightarrow |d\rangle$, and $|d\rangle \rightarrow -|b\rangle$, so that $|aa\rangle \rightarrow -|cc\rangle$, etc. As a consequence, the cross sections $\sigma_{ac,a \rightarrow a}$ and $\sigma_{ac,c \rightarrow c}$ are complex conjugated, as are $\sigma_{ac,b \rightarrow b}$ and $\sigma_{ac,d \rightarrow d}$, while $\sigma_{ac,a \rightarrow c}$ and $\sigma_{ac,c \rightarrow a}$ are real. By the same symmetry argument $\sigma_3^{(\text{DIS})} = 0$.

B. First-order correction

The vanishing values of λ_1 and λ_2 for degenerate internal states prompt us to resort to a more rigorous approach for gaining insight into the $B \neq 0$ frequency stability. This leads us to calculate first-order corrections in the hyperfine level splittings. As pointed out in Refs. 7 and 8, these cannot be calculated by straightforward first-order perturbation theory, i.e., the Born approximation: The perturbation $V^{\text{hf}} + V^Z - \bar{\epsilon}$, in which $\bar{\epsilon}$ is the average internal energy, does not fall off with interatomic distance, so that distorted-wave Born integrals do not converge. We have shown that a first-order method previously devised for nuclear reactions¹⁶ can also be applied successfully to individual partial waves in sub-Kelvin H+H elastic scattering. In this paper we have to deal also with inelastic S -matrix elements, i.e., we would like to dispose of a first-order correction to the above-mentioned DIS approximation. The following elegant expression can be derived:¹⁷

$$\Delta S_{\{\alpha'\beta'\},\{\alpha\beta\}}^l = (k_{\alpha'\beta'} k_{\alpha\beta} / \bar{k}^2)^{l+1/2} \times \langle \{\alpha'\beta'\} | (P_T - P_S) (V^{\text{hf}} + V^Z - \bar{\epsilon}) \times (P_T - P_S) | \{\alpha\beta\} \rangle \Delta^l(\bar{k}), \quad (16)$$

where

$$\Delta^l(\bar{k}) = \frac{i}{4} \frac{m_H}{\hbar} \left[\int_0^{r_0} (u_T^{l(0)} - u_S^{l(0)})^2 dr + \frac{1}{2\bar{k}} (S_T^{l(0)} - S_S^{l(0)})^2 \times W_r \left[O_l(\bar{k}, r), \frac{\partial}{\partial \bar{k}} O_l(\bar{k}, r) \right] \right]. \quad (17)$$

For the notation we refer to Refs. 7 and 8. The integral term in Eq. (17) is the Born-type integral that would have been obtained with a perturbation $V^{\text{hf}} + V^Z - \bar{\epsilon}$ confined to a sphere with radius r_0 in relative orbital space, enclosing the range of the central interaction $V^c(r)$. The Wronskian surface term takes into account the effect of the perturbation outside this sphere.

As an example we give the expression for one of the inelastic S -matrix elements, corresponding to the zeroth-order equation (12),

$$\Delta S_{aa,\{ac\}}^l = (k_{aa} k_{ac} / \bar{k}^2)^{l+1/2} \frac{1}{\sqrt{2}} \times \sin 4\theta \frac{2\epsilon_a - \epsilon_b - \epsilon_d}{\hbar} \Delta^l(\bar{k}). \quad (18)$$

Making use of such expressions we find the hyperfine-plus-Zeeman-induced frequency-shift parameters

$$\begin{aligned} \Delta\lambda_0 &= \frac{\pi a}{\hbar k^2} \sum_{l \text{ even}} (2l+1) \left[\left(\frac{1}{2} - \frac{3}{2} \cos^2 2\theta \right) \text{Im} \xi_T^l - \frac{1}{4} \sin 2\theta \cos^2 2\theta \text{Im} \eta_-^l \right], \\ \Delta\lambda_1 &= \frac{\pi a}{\hbar k^2} \sum_{l \text{ odd}} (2l+1) \frac{1}{2} \sin 2\theta \text{Im} (\xi_T^l + \xi_S^l) \\ &\quad - \frac{\pi a}{\hbar k^2} \sum_{l \text{ even}} (2l+1) (\sin 2\theta \text{Im} \xi_T^l + \frac{1}{4} \sin 2\theta \cos^2 2\theta \text{Im} \eta_+^l), \\ \Delta\lambda_2 &= -\frac{\pi a}{\hbar k^2} \sum_{l \text{ odd}} (2l+1) \frac{1}{2} \sin 2\theta \text{Im} (\xi_T^l + \xi_S^l), \\ \Delta\lambda_3 &= 0, \end{aligned} \quad (19)$$

$$\begin{aligned} \Delta\sigma_0 &= \frac{\pi a}{\hbar k^2} \sum_{l \text{ even}} (2l+1) \sin 2\theta \\ &\quad \times \{ \text{Re} \xi_T^l - \frac{1}{4} \cos^2 2\theta \\ &\quad \times \text{Re} [2(\xi_T^l - \xi_S^l) + \eta_-^l] \} \end{aligned}$$

$$\begin{aligned} \Delta\sigma_1 &= -\frac{\pi a}{\hbar k^2} \sum_{l \text{ even}} (2l+1) \left[\left(\frac{1}{2} + \frac{1}{2} \cos^2 2\theta \right) \text{Re} \xi_T^l \right. \\ &\quad \left. + \frac{1}{16} \sin^2 4\theta \text{Re} \eta_+^l \right], \\ \Delta\sigma_2 &= 0, \end{aligned}$$

$$\Delta\sigma_3 = -\frac{\pi a}{\hbar k^2} \sum_{l \text{ odd}} (2l+1) \frac{1}{4} \sin 4\theta \text{Re} (\xi_T^l - \xi_S^l),$$

in which the shorthand notation

$$\xi_T^l(k) = \Delta^{l*}(k) e^{2i\delta_T^l(k)} \quad (20)$$

has been introduced for triplet scattering and similarly for the singlet case, while

$$\begin{aligned} \eta_+^l(k) &= (kk_> / \bar{k}^2)^{2l+1} [\xi_T^l(\bar{k}_>) - \xi_S^l(\bar{k}_>)] \\ &\quad + (kk_< / \bar{k}^2)^{2l+1} [\xi_T^l(\bar{k}_<) - \xi_S^l(\bar{k}_<)] \\ &\quad - 2[\xi_T^l(k) - \xi_S^l(k)], \\ \eta_-^l(k) &= (kk_> / \bar{k}^2)^{2l+1} [\xi_T^l(\bar{k}_>) - \xi_S^l(\bar{k}_<)] \\ &\quad - (kk_< / \bar{k}^2)^{2l+1} [\xi_T^l(\bar{k}_<) - \xi_S^l(\bar{k}_<)]. \end{aligned} \quad (21)$$

IV. NUMERICAL RESULTS AND CONSEQUENCES FOR H MASER

The introduction of a strong B field has a number of consequences for the operation of the hydrogen maser which are associated with the dependence of hyperfine spin functions and level splittings on B . In the first place the coupling of the a and c levels due to the interaction with the magnetic field of the radiation mode varies with B . As a consequence, B has a direct influence on the maser operation and in particular on the oscillation condition. This can be derived in complete analogy to the $B=0$ case.¹⁸ The result is

$$\frac{1}{T_1 T_2} < \frac{n_H V_b Q_c \mu_0 \mu_e^2 \eta (\rho_{cc} - \rho_{aa})_0 \sin^2 2\theta}{\hbar V_c T_b} \sin^2 2\theta, \quad (22)$$

where $(\rho_{cc} - \rho_{aa})_0$ characterizes the substate populations for the H atoms entering the cavity. The further notation follows Ref. 18.

A second effect, on which we have concentrated already in the foregoing, is the influence of B on H+H collisions and consequently on hyperfine population dynamics and collisional frequency shift. Elastic S -matrix elements, which already determine this frequency shift for $B=0$, change considerably by the introduction of a B field of, for instance, 0.05 T. In addition, significant contributions from inelastic transitions are introduced.

We evaluated the above-mentioned DIS frequency parameters corrected to first order for the hyperfine level splitting. The total parameters have also been calculated rigorously using the coupled-channel method. Deviations are expected to occur close to and below inelastic thresholds, for instance, in the aa channel due to the cc channel. As for $B=0$,⁷ discontinuities due to these thresholds are described by the coupled-channel method, but not by the DIS approximation plus first-order correction. Despite these shortcomings, the DIS plus first-order method is very useful because it leads to more explicit expressions, especially with respect to the B dependence, which facilitates qualitative insight. In addition, it serves as an important test case for the coupled-channel calculations.

In Fig. 1 we present the λ and σ parameters as a function of B at a fixed collision energy of 0.6 K, as predicted by the coupled-channel calculations. It turns out that the field dependence is remarkably accurately (to the percent level relative to the values at the maxima) described by the simple DIS plus first-order polynomials in $\sin 2\theta$ and $\cos 2\theta$ of the expressions (14) and (19). In particular, the $\sin 2\theta$ proportionality of λ_1 and λ_2 is fully confirmed, as well as the $1 + \cos^2 2\theta$ dependence of σ_1 and σ_2 . It is also of interest to point to the change of sign of λ_0 at $B=35.9$ mT, due to its $1 - 3 \cos^2 2\theta$ field dependence. At this field strength the spin-exchange tuned cavity frequency is expected to be equal to the atomic frequency. We note also that λ_0 , λ_3 , σ_1 , and σ_2 are dominated by their DIS contributions and λ_1 , λ_2 , and σ_3 by their first-order parts. The σ_0 cross section receives significant contributions from both zeroth- and first-order parts.

The same characteristics are found at higher collision energies. As pointed out previously, for lower collision

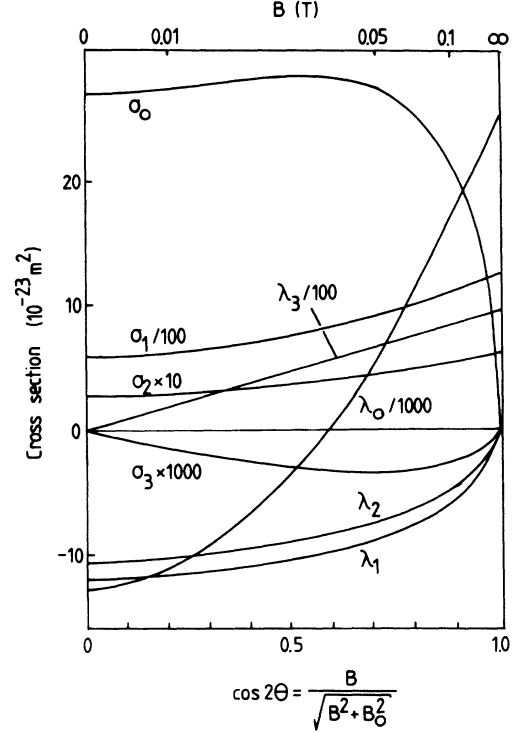


FIG. 1. Magnetic field dependence of frequency-shift parameters for fixed collision energy 0.6 K.

energies pronounced deviations occur due to thresholds in inelastically coupled channels. The Boltzmann averaged frequency shift parameters at the relevant temperatures $T=0.52$ K and higher are, however, much less sensitive to such deviations. Using the simple field dependences, the $\bar{\lambda}_i$ and $\bar{\sigma}_i$ parameters may be expressed simply in their previously given values^{7,8} for $B=0$, except for $\bar{\sigma}_0$, $\bar{\lambda}_3$, and $\bar{\sigma}_3$. The latter two are “new” parameters, which vanish at $B=0$. In Fig. 2 we give them at $B=B_0 \approx 50.7$ mT. Values for other fields may be obtained from their field dependences $\cos 2\theta$ and $\sin 4\theta$, respectively. Furthermore, $\bar{\sigma}_0$ can be expressed linearly in terms of its previously given values at $B=0$ and those at $B=B_0$,

$$\begin{aligned} \bar{\sigma}_0(B, T) = & \bar{\sigma}_0(0, T) \sin 2\theta (1 - 2 \cos^2 2\theta) \\ & + \bar{\sigma}_0(B_0, T) 2\sqrt{2} \sin 2\theta \cos^2 2\theta. \end{aligned} \quad (23)$$

In Fig. 2 we also present $\bar{\sigma}_0(B_0, T)$.

We now turn to the implications of a stronger B field for the frequency stability. With present experimental possibilities in connection with hyperfine state selection in H masers it is possible to inject equal populations of the b and d hyperfine levels into the storage bulb. Making use of conservation of the total electronic plus nuclear spin projection along \mathbf{B} , one thus expects that the $\rho_{dd} - \rho_{bb}$ term in the frequency shift can be sufficiently eliminated. We are then left with a collisional frequency shift of the form

$$\delta\omega_c = n_H \langle v \rangle [(\rho_{cc} - \rho_{aa}) \bar{\lambda}_0 + (\rho_{cc} + \rho_{aa}) \bar{\lambda}_1 + \bar{\lambda}_2], \quad (24)$$

with coefficients $\bar{\lambda}_i(B, T)$. From a self-consistent calcula-

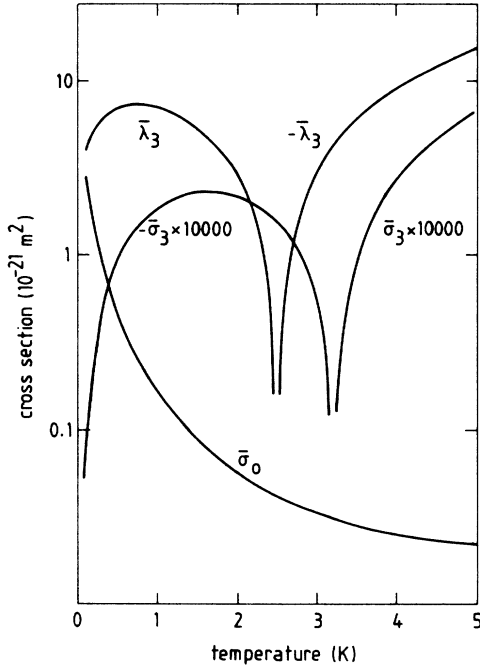


FIG. 2. Temperature dependence of $\bar{\sigma}_0$, $\bar{\lambda}_3$, and $\bar{\sigma}_3$ for fixed magnetic field $B_0 \approx 50.7$ mT.

tion of the H-maser oscillation similar to that for $B=0$, we again find $\rho_{cc} - \rho_{aa}$ to be proportional to the transverse relaxation rate

$$\rho_{cc} - \rho_{aa} = \frac{\hbar V_c (1 + \Delta^2)}{m_H \mu_0 (\mu_e + \mu_p)^2 \eta Q_c V_b \sin^2 2\theta} T_2^{-1}, \quad (25)$$

which again contains a B -dependent $\sin^2 2\theta$ factor. For the further notation we refer to Ref. 7. It follows that the spin-exchange cavity tuning procedure can still be used to eliminate the $\rho_{cc} - \rho_{aa}$ term of the frequency shift. On the basis of the usual very weak magnetic field the remaining frequency shift could not be eliminated by a similar cavity tuning procedure. For the prospects for $B \neq 0$ to be more favorable, it should be possible to write the shift as a linear function of

$$T_2^{-1} = (T_2^{-1})_0 + n_H \langle v \rangle [\bar{\sigma}_1 (\rho_{cc} + \rho_{aa}) + \bar{\sigma}_2]. \quad (26)$$

In this equation we have left out a negligible $\bar{\sigma}_0 (\rho_{cc} - \rho_{aa})$ term. Clearly, the remaining collisional frequency shift is insensitive to experimental fluctuations in n_H if the dimensionless frequency-shift parameter

$$\Omega = - \frac{\bar{\lambda}_1 (\rho_{cc} + \rho_{aa}) + \bar{\lambda}_2}{\bar{\sigma}_1 (\rho_{cc} + \rho_{aa}) + \bar{\sigma}_2} \quad (27)$$

is zero or at least independent of $\rho_{cc} + \rho_{aa}$.

For $B \neq 0$ it follows from Eq. (14) that Ω is again a hyperfine splitting induced quantity. From Fig. 1 and Refs. 7 and 8 it follows that $\bar{\lambda}_1$ and $\bar{\lambda}_2$ are both negative and $\bar{\sigma}_1$ and $\bar{\sigma}_2$ both positive for all reasonable field strengths, so that a value zero for Ω cannot be expected to be achieved. In fact, because of the similar field dependences of λ_1 and λ_2 , and of σ_1 and σ_2 , Ω depends on B via a $\sin 2\theta / (1 + \cos^2 2\theta)$ overall factor.

The next-best possibility, an Ω value independent of $\rho_{cc} + \rho_{aa}$, would be possible when for certain B the ratio $\bar{\lambda}_2 / \bar{\lambda}_1$ would be equal to $\bar{\sigma}_2 / \bar{\sigma}_1$ at $T = 0.52$ K. Since these ratios are in good approximation field independent, this weaker condition is already excluded by our previous $B=0$ results: $\bar{\sigma}_2 / \bar{\sigma}_1$ is at least two orders of magnitude smaller than $\bar{\lambda}_2 / \bar{\lambda}_1$. We conclude that a stronger B field does not create new possibilities to eliminate the frequency instability associated with fluctuations in n_H .

V. CONCLUSIONS

The operation of the (cryogenic) H maser, especially its recent recirculating version, depends on a complicated interplay of hyperfine level occupations and coherences. A valuable source of information on these quantities is the maser oscillation frequency. From its measured value as a function of experimental parameters such as cavity frequency and atomic flux, and using its dependence on level populations it is possible to gain information on the population dynamics. From this point of view it would seem very useful to introduce an external constant magnetic field as an additional experimental parameter to diagnose the population dynamics. In this paper we have predicted the theoretical B dependence of the maser oscillation frequency needed for the above-mentioned analysis. We have derived a $B \neq 0$ expression for the collisional frequency shift in terms of hyperfine level populations. The coefficients $\bar{\lambda}_i$ in this expression, as well as the corresponding quantities $\bar{\sigma}_i$ determining the transverse relaxation rate, have been calculated in zeroth and first order in the hyperfine level splittings, as well as on the basis of the rigorous coupled-channel method.

A second context in which our results might be useful is associated with a possible use of the (cryogenic) hydrogen maser as a precision instrument for measuring specific phenomena in a dilute quantum gas. Insofar as an external magnetic field is essential for such effects, for instance, in the case of nuclear or electronic spin waves in atomic hydrogen, it is essential to understand the influence of a B field on the operation of the H maser.

Finally, we have discussed the implications of our calculated frequency-shift parameters for the frequency stability of the cryogenic H maser. We find that the introduction of a stronger B field does not eliminate the source of frequency instabilities pointed out previously for the conventional setup based on a very weak B field.

¹H. M. Goldenberg, D. Kleppner, and N. F. Ramsey, Phys. Rev. Lett. **5**, 361 (1960).

²S. B. Crampton, W. D. Phillips, and D. Kleppner, Bull. Am. Phys. Soc. **23**, 86 (1978).

³R. F. C. Vessot, M. W. Levine, and E. M. Mattison, NASA Technical Memorandum No. 78104, 1978 (unpublished), p. 549.

⁴M. Morrow, R. Jochemsen, A. J. Berlinsky, and W. N. Hardy,

- Phys. Rev. Lett. **46**, 195 (1981).
- ⁵A. J. Berlinsky and W. N. Hardy, NASA Conference Publication No. 2220, 1982 (unpublished), p. 547.
- ⁶L. C. Balling, R. J. Hanson, and F. M. Pipkin, Phys. Rev. **133**, A607 (1964); **135**, AB1 (1964); P. L. Bender, *ibid.* **132**, 2154 (1963); A. C. Allison, Phys. Rev. A **5**, 2695 (1972); A. J. Berlinsky and B. Shizgal, Can. J. Phys. **58**, 881 (1980).
- ⁷B. J. Verhaar, J. M. V. A. Koelman, H. T. C. Stoof, O. J. Luiten, and S. B. Crampton, Phys. Rev. A **35**, 3825 (1987).
- ⁸J. M. V. A. Koelman, S. B. Crampton, H. T. C. Stoof, O. J. Luiten, and B. J. Verhaar, Phys. Rev. A **38**, 3535 (1988).
- ⁹M. D. Hürlimann, W. N. Hardy, M. E. Hayden, and R. W. Cline, in *Proceedings of the Fourth Symposium on Frequency Standards and Metrology, Ancona, 1988*, edited by A. De Marchi, Springer-Verlag, Berlin, 1989), p. 95; W. N. Hardy, M. D. Hürlimann, and R. W. Cline, Jap. J. of Appl. Phys. **26**, 2065 (1987).
- ¹⁰I. F. Silvera, R. F. C. Vessot, and R. L. Walsworth (private communication).
- ¹¹E. P. Bashkin, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 320 (1989) [JETP Lett. **49**, 363 (1989)]; (private communication).
- ¹²H. T. C. Stoof, J. M. V. A. Koelman, and B. J. Verhaar, Phys. Rev. B **38**, 4688 (1988).
- ¹³J. M. V. A. Koelman, H. T. C. Stoof, B. J. Verhaar, and J. T. M. Walraven, Phys. Rev. Lett. **59**, 676 (1987).
- ¹⁴C. C. Agosta, I. F. Silvera, H. T. C. Stoof, and B. J. Verhaar, Phys. Rev. Lett. **62**, 2361 (1989).
- ¹⁵E. Tiesinga, H. T. C. Stoof, and B. J. Verhaar (unpublished).
- ¹⁶A. M. Schulte and B. J. Verhaar, Nucl. Phys. **A232**, 215 (1974).
- ¹⁷A. C. Maan, H. T. C. Stoof, and B. J. Verhaar (unpublished).
- ¹⁸D. 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); S. B. Crampton, *ibid.* **158**, 57 (1967).