Surface-state hydrogen maser

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We describe a hydrogen maser operating at very low temperatures in which most of the hydrogen atoms are condensed on a superfluid helium surface in long-lived states. This proposed maser can be used to obtain information on the properties of the hydrogen−liquid-helium-surface system. In addition, it promises to be an interesting system from the point of view of nonlinear dynamics. It is found that the surface recombination to molecular hydrogen, which might be considered as undesirable, is actually necessary to achieve the masing conditions. We develop the maser equations and consider a number of realistic conditions for operation.

I. INTRODUCTION

In this paper we propose an alternative type of maser, the surface cryogenic hydrogen maser (SCHM), in which the dominant species coupled to the radiation field are the hydrogen atoms condensed on a superfluid helium surface. The SCHM provides a method for studying hydrogen-surface interactions, as well as an interesting device to study nonlinear dynamic behavior.

Our analysis treats atoms in the volume and surface states on an equal footing, using the Maxwell-Bloch equations coupled with the rate equations for the density of the hydrogen hyperfine states. We find the unexpected result that recombination of the hydrogen atoms is required to satisfy the masing condition. We analyze a "conventional" mode of operation in which the masing takes place on the a-to-c transition, shown in the hyperfine diagram (Fig. 1). In addition, we consider the unusual operation on the a-to-b transition in which the population inversion is produced not only by the incoming beam, but also by surface recombination of hydrogen in the cavity. Operating conditions are presented in both cases. Before entering into details of the analysis we present some introductory discussion.

The room-temperature atomic-hydrogen maser is the most stable time standard currently available for measurement intervals between 1 and 10^4 s. More recently a sub-Kelvin version, called a cryogenic hydrogen maser (CHM), has been proposed [1] and constructed [2]. The CHM works in essentially the same manner as the room-temperature maser, but has a number of advantages due to the low temperature which promise stability greatly enhanced over that of the room-temperature maser. The CHM involves atoms in the cavity volume coupled to the radiation field, but perturbed by the helium surface, resulting in a wall frequency shift which depends on temperature as well as the area-to-volume ratio. The CHM operates at a temperature of about 500 mK. The SCHM takes advantage of the H−liquid-helium adsorption iso-

therm. By cooling to the 50−100-mK temperature range H atoms populate long-lived surface states. In this regime we find conditions in which the surface plays a more prominent role than that of perturbing the effective volume parameters. In this connection one could think of a situation in which the pulse time in a pulsed oscillation regime [3] is so short that it becomes of the order of the average sticking time of atoms to the surface. In this case surface and volume atoms could have different time-dependent spin behaviors and by enhancing the number of atoms at the surface relative to those in the volume, the surface-state atoms dominate the maser
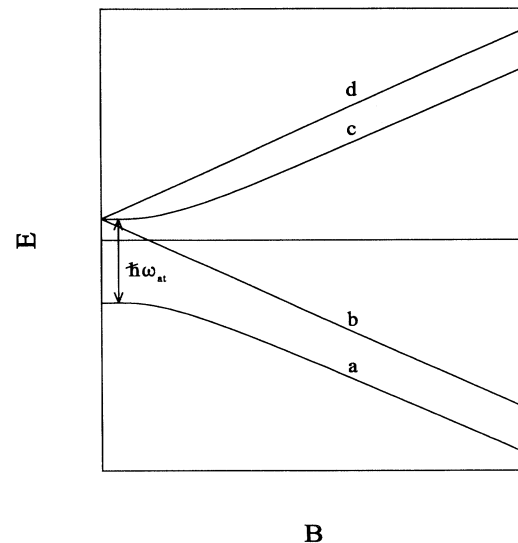

FIG. 1. Schematic diagram of the energy of the four hyperfine levels of the hydrogen $1s$ ground state as a function of the applied magnetic field. In the "conventional" maser oscillation takes place between the a and c levels, indicated with an arrow. For the SCHM the a-b transition is also considered.

behavior. Note that due to the sensitivity to perturbations in between pulses the exact time-dependent spin behavior of surface atoms could already be important for the operation of the CHM in the pulsed regime.

A second motivation for the present research is the possibility to use the dependence of the maser operation on the surface for precision measurements. Partially, this has already been done in CHM experiments in which it has been possible to determine the transition frequency of hydrogen atoms adsorbed at the superfluid-helium surface with great accuracy [4]. One can imagine that in a maser in which the dominant species coupled to the radiation field are hydrogen atoms condensed at the surface, much more information can be obtained about hydrogen-surface interactions, e.g., an alternative, possibly more accurate, determination of the surface adsorption energy and the influence of dipolar collisions and recombination on the atomic linewidth.

A third motivation for the investigations is the possibility to use the transition frequency of hydrogen atoms adsorbed at the helium surface as the basis for a primary time standard. The wall shift inhibits the use of present hydrogen masers as a primary time standard. In a SCHM one would obviously have to deal with a volume shift. Whereas the wall shift cannot be eliminated in the CHM, there is every reason to hope that a volume shift can be made arbitrarily small by keeping the atoms at the surface longer, e.g., by lowering the temperature.

In the following, we first generalize the conventional Maxwell-Bloch equations to a combined set of equations for volume and surface in Sec. II. In Sec. III we show how the usual maser equations and maser parameters follow from the generalized Maxwell-Bloch equations in the limit of small sticking times. In Sec. IV we concentrate on new phenomena both for the traditional scheme in which oscillation takes place between the a and c levels of the hydrogen ground state and for a maser that oscillates between the a and b levels. In Sec. V we consider some aspects that are related to the experimental realization of the SCHM. Some conclusions are presented in Sec. VI.

II. GENERALIZED MAXWELL-BLOCH EQUATIONS

The usual way to describe the dynamics of the hydrogen maser is by using the Maxwell-Bloch equations

$$\dot{B} = -(\kappa + i\omega_c)B + gM \; ,$$
$$\dot{M} = -(\gamma_\perp + i\omega_{at})M + gB\Delta \; ,$$
$$\dot{\Delta} = -\gamma_\parallel(\Delta - \Delta_0) - 2g(BM^* + B^*M) \tag{1}$$

for the complex magnetic field B, the complex magnetization M, and the population inversion Δ. The field B is normalized so as to equal the square root of the number of photons, whereas $M = N\rho_{ca}$ and $\Delta = N(\rho_{cc} - \rho_{aa})$ in terms of the one-atom spin-density matrix ρ and the number N of atoms in the storage bulb. The subscripts of ρ stand for the one-atom hyperfine states. The cavity resonance frequency is denoted by ω_c, the atomic transition frequency by ω_{at}, the cavity damping rate by

$\kappa = \omega_c / 2Q_c = 1/T_c$, with Q_c the quality factor of the cavity, and the damping rates for M and Δ by $\gamma_\perp = 1/T_2$ and $\gamma_\parallel = 1/T_1$, the transverse and longitudinal relaxation rates. The equilibrium value of Δ in the absence of an atom-field interaction is denoted by Δ_0. Finally, the one-photon Rabi frequency g is given by

$$g^2 = \frac{\mu_0(\mu_e + \mu_p)^2\eta\omega_c}{2\hbar V_c} \; , \tag{2}$$

with V_c the cavity volume, μ_0 the vacuum permeability, μ_e (μ_p) the electron (proton) magnetic moment, and η the filling factor.

The atomic transition frequency ω_{at} and the transverse and longitudinal relaxation rates $\gamma_\perp$ and $\gamma_\parallel$ entering Eqs. (1) differ from the values for an unperturbed hydrogen atom due to collisions with other hydrogen atoms and helium atoms and due to the finite residence time in the cavity field. Another effect which influences the frequency and the width of the atomic transition, and which is the primary interest of this paper, is the sticking of atoms to the wall of the storage bulb. The atoms traverse the storage bulb many times before leaving it. Each time an atom hits the wall it has a finite probability to stick. In the case of a sticking event it will desorb after an average time τ_s. The average time τ_s of a sticking event and the average time τ_v between subsequent stickings are given by

$$\frac{1}{\tau_s} = \frac{kT}{2\pi\hbar}se^{-E_b/kT} \; ,$$
$$\frac{1}{\tau_v} = \left[\frac{kT}{2\pi m_H}\right]^{1/2}\frac{A_b}{V_b}s \; , \tag{3}$$

where k is Boltzmann's constant, T is the temperature, s ($=0.33$ K^{-1} T) is the sticking probability, and E_b is the binding energy of the hydrogen atoms to the superfluid ^{4}He surface ($E_b = 1.0$ K). The mass of the hydrogen atom is denoted by m_H whereas the surface area of the storage bulb is given by A_b and its volume by V_b. The energy levels of a hydrogen atom bound to the surface will be perturbed due to the interaction with the helium surface, which results in a shift of the transition frequency. This means that the transition frequency has two different values, one in the volume and one at the surface.

The net phase effect of two alternating transition frequencies has been discussed by Anderson and Weiss [5] and applied to the hydrogen maser by Morrow and Hardy [6]. The phase shift which the atoms undergo per sticking event due to the difference in atomic transition frequencies is calculated assuming that the sticking time and the time between stickings are distributed according to Poisson statistics. In the usual situation for the cold-hydrogen maser the result takes on a particularly simple form. The rapid exchange of atoms among volume and surface states and the very short sticking time compared to the time in the volume result in a shift (wall shift) of the volume frequency due to the influence of the transition frequency at the surface and it is not necessary to deal with the surface atoms explicitly. Consequently, the Maxwell-Bloch equations (1) remain valid but with

effective values for ω_{at} and $\gamma_\perp$:

$$\omega_{at}=\omega_v+\frac{1}{\tau_v}\frac{\phi_0}{1+\phi_0^2} \; ,$$

$$\gamma_\perp=\gamma_\perp^v+\frac{1}{\tau_v}\frac{\phi_0^2}{1+\phi_0^2} \; ,$$

(4)

where ω_v and $\gamma_\perp^v$ are the atomic transition frequency and transverse relaxation rate in the volume and $\phi_0=\tau_s(\omega_s-\omega_v)$ is the average phase shift per sticking event, with ω_s being the atomic transition frequency at the surface.

This approach is valid as long as there is a rapid exchange of atoms between surface and volume and the total time spent at the surface is short compared to the time spent in the volume. This approach also neglects recombination and relaxation effects and any independent interaction with the radiation field during the time that the atoms reside at the surface.

Generalizing the usual derivation [3] of Eqs. (1) to the case of a combined system of atoms at the surface and in the volume, we obtain

$$\dot{B}=-(\kappa+i\omega_c)B+g(M^v+M^s) \; ,$$

$$\dot{M}^v=-(\gamma_\perp^v+i\omega_v)M^v+gB\Delta^v-\frac{1}{\tau_v}M^v+\frac{1}{\tau_s}M^s \; ,$$

$$\dot{M}^s=-(\gamma_\perp^s+i\omega_s)M^s+gB\Delta^s+\frac{1}{\tau_v}M^v-\frac{1}{\tau_s}M^s \; ,$$

(5)

$$\dot{\Delta}^v=-\gamma_\parallel^v(\Delta^v-\Delta_0^v)-2g(BM^{v*}+B^*M^v)$$
$$-\frac{1}{\tau_v}\Delta^v+\frac{1}{\tau_s}\Delta^s \; ,$$

$$\dot{\Delta}^s=-\gamma_\parallel^s(\Delta^s-\Delta_0^s)-2g(BM^{s*}+B^*M^s)$$
$$+\frac{1}{\tau_v}\Delta^v-\frac{1}{\tau_s}\Delta^s \; ,$$

with separate quantities M^i and Δ^i ($i=v,s$) for volume and surface. The γ coefficients are the transverse and longitudinal relaxation rates in the volume and at the surface and the Δ_0 coefficients stand for the equilibrium values of the population inversion in the absence of atom exchange and atom-field interaction.

In order to evaluate possible operating conditions for the SCHM knowledge of the various constants in Eqs. (5) is essential. As far as the operation of the hydrogen maser depends on specific device parameters we have assumed values which correspond to the Harvard-Smithsonian CHM [2], except for the area-to-volume ratio of the storage bulb, which we consider to be a free parameter. In view of the operational possibilities of the surface maser it is necessary to increase this considerably compared to the volume maser. This is a result following from the simulation to be described in Sec. IV. Such an increase can be accomplished by including in the storage volume a porous material or a configuration of plates analogous to that used in Ref. [7].

The relaxation rates in the volume describe the loss of coherence and population inversion primarily due to

atoms leaving the storage bulb and to spin-exchange collisions. We neglect the hyperfine-induced contribution to $\gamma_\perp^v$. Specifically, $\gamma_\parallel^v=\gamma_b+n_v\langle v_v\rangle\bar{\sigma}$ and $\gamma_\perp^v=\gamma_b+\frac{1}{2}n_v\langle v_v\rangle\bar{\sigma}$, where γ_b is the rate at which atoms leave the storage bulb, n_v is the density of atoms in the volume, $\langle v_v\rangle$ is the thermal velocity of atoms in the volume, and $\bar{\sigma}\equiv\langle v_v\sigma_c\rangle/\langle v_v\rangle$, with σ_c the spin-exchange cross section. We take γ_b to be proportional to $\sqrt{T}$ with the value 0.2 s^{-1} at $T=0.5$ K and for $\bar{\sigma}$ we take a constant value of 8×10^{-17} cm^2 [8] in the temperature range under consideration, using $\rho_{cc}+\rho_{aa}=\rho_{bb}+\rho_{dd}=\frac{1}{2}$. The equilibrium value of the population inversion in the volume is determined by the influx of atoms into the storage bulb and by collisions, $\gamma_\parallel^v\Delta_0^v=I(\rho_{cc}-\rho_{aa})_{entr}+n_v\langle v_v\rangle\bar{\sigma}\Delta_c^v$, with I the flux of atoms into the storage bulb, $(\rho_{cc}-\rho_{aa})_{entr}=\frac{1}{2}$ the population inversion of these atoms, and Δ_c^v determined by the thermal Boltzmann distribution.

As the direct inflow and outflow of atoms is absent at the surface, the relaxation rates and the equilibrium value of the population inversion at the surface are primarily determined by collisions. In analogy with the volume parameters we have $\gamma_\parallel^s=\sigma\langle v_s\rangle\bar{l}$ and $\gamma_\perp^s=\frac{1}{2}\sigma\langle v_s\rangle\bar{l}$, with σ the number of atoms at the surface per cm^2, $\langle v_s\rangle$ the thermal velocity of atoms at the surface, and $\bar{l}$ the thermally averaged collisional cross length. We take this cross length as a sum of contributions from recombination and from two-body scattering due to the spin-exchange potential and the dipole-dipole interaction. For the recombination contribution to $\bar{l}$ we take the value 3×10^{-9} cm [9]. Calculations for the spin-exchange contribution neglecting motion perpendicular to the surface [10] and order of magnitude estimates for the dipolar contribution [9,11] indicate that these are at most equally important. The product $\gamma_\parallel^s\Delta_0^s$ is a sum of $\gamma\Delta$ products from the above processes. The Δ value associated with recombination vanishes, while the spin-exchange and dipole-dipole contributions have Boltzmann equilibrium values. It should be noted that the above-mentioned parameter choices are partly based on order-of-magnitude estimates. The existing theoretical description of two-body exchange and dipolar collisions and two-body recombination at the surface is still rather incomplete. The determination of actual experimental SCHM operating parameters will be a very welcome addition to the available knowledge of the properties of the hydrogen-surface system.

In the a-c maser both Δ_0^v and Δ_0^s decrease due to collisions. In a maser based on the a-to-b transition, however, one has the interesting situation that preferential relaxation and recombination of the a-state atoms increase the population inversion, which facilitates maser oscillation. This effect will be dealt with in Sec. IV. The remaining parameters in Eqs. (5) are of the same order of magnitude for the a-b maser as for the a-c maser.

Note that in the derivation of Eqs. (5) it was tacitly assumed that the total number of atoms in the volume (N_v) and at the surface (N_s) are constant. If this restriction is relaxed, Eqs. (5) have to be supplemented with additional equations for the time dependence of N_v and N_s.

Taking into account the persistent problem from dipolar relaxation in experiments with hydrogen on a helium film, one would expect that the dipolar interaction would also affect the performance of a surface maser drastically. One should, however, keep in mind some differences with the more usual situation of gas samples which are almost completely doubly polarized (H↓‡ or H↑‡). Whereas for the latter dipolar relaxation dominates over the direct surface recombination, the situation is different in both the a-c and a-b surface maser because of the large fraction of a-state atoms. Two different processes have to be distinguished, i.e., the loss of coherence and population inversion due to hyperfine-state-changing collisions and the loss of coherence due to the phase which the oscillating magnetic dipole moment acquires during an elastic dipolar collision. Since we are considering zero magnetic field, the dipolar interactions are dominated by the electron spins. This might suggest a large contribution to $\gamma_\parallel^s$ and $\gamma_\perp^s$. However, at zero magnetic field also the recombination rate is large. The magnitude of the dipolar T_1^{-1} and T_2^{-1} due to hyperfine-state-changing collisions turns out to be negligible compared to the recombination contribution [9] under the conditions considered $[\rho_{cc}(\rho_{bb}) \simeq \rho_{aa}]$. This is not necessarily true for the second process mentioned above, i.e., the change of phase of the oscillating magnetic dipole moment in the magnetic field of a neighboring atom. However, in the case of the a-b maser one can choose a setup based on including in the storage volume a configuration of very smooth plates analogous to that used in Ref. [7], so that variations in the orientation of the surface normal would be negligible, producing a single atomic transition frequency at the surface. This possibility was already mentioned earlier in this section. The possibility of a porous medium, which was also mentioned, is excluded. The plate configuration would reduce the dipolar width of the order of 10^5–10^6 Hz, observed in Ref. [11], by the 10^5-fold-lower surface densities (see below) to a value of, at most, the same order as the recombination T_2^{-1}, given above. In the case of the a-c maser an average dipolar field would be absent if $\rho_{bb} = \rho_{dd}$. To the extent that this equality is not satisfied one may use a smooth plate configuration also in this case.

III. INFLUENCE OF THE SURFACE FOR SHORT STICKING TIMES

Under the usual operating conditions of the sub-Kelvin hydrogen maser, the atoms spend most of their time in the volume and τ_s is by far the shortest time constant in Eqs. (5). A treatment in which τ_s is considered as a small parameter therefore seems to be appropriate. However, even for short sticking times the dephasing at the surface can be considerable due to the large difference between ω_v and ω_s. In view of this we eliminate a rapid time dependence $\exp(-i\omega_v t)$, introduce the reduced time $\bar{t} = t/\tau_s$, and transform to quantities $\bar{M}$ and $\bar{\Delta}$, which can take on the maximum value 1,

$$\bar{B} = B e^{i\omega_v t} \ , \quad \bar{M}^i = M^i e^{i\omega_v t}/N_i \ , \quad \bar{\Delta}^i = \Delta^i/N_i$$
$$(i = v, s) \ , \quad (6)$$

with $N_s/N_v = \tau_s/\tau_v$. By a standard two-times approach [12] we find the rapid changes by leaving out all terms containing τ_s, except ϕ_0. On the time scale τ_s we thus find that $\bar{B}$, $\bar{M}^v$, and $\bar{\Delta}^v$ are constant, while $\bar{M}^s$ and $\bar{\Delta}^s$ decay rapidly to the values

$$\bar{M}^s = \frac{1}{1 + i\phi_0} \bar{M}^v \ , \quad \bar{\Delta}^s = \bar{\Delta}^v \ . \quad (7)$$

Substituting these values in the terms of order τ_s we find on a longer time scale the usual maser equations (1) with precisely the effective atomic frequency and transverse relaxation rates (4), found usually by the more complicated derivation in Refs. [5,6].

IV. PROSPECTS FOR CONSTRUCTING A SURFACE MASER

The description in which the surface magnetization and population inversion adiabatically follow their volume counterparts requires sticking times that are short compared to other characteristic times appearing in Eqs. (5), such as $1/\kappa$, $1/\gamma_\parallel^v$, $1/\gamma_\parallel^s$, $1/\gamma_\perp^v$, $1/\gamma_\perp^s$, $1/gB$, and τ_v. If, however, the temperature of the maser is lowered considerably below present values, τ_s becomes of the order of, or even larger than, a number of these time constants. The approach of the preceding section is then not justified and the dynamics of the surface has to be included in the description of the hydrogen maser.

As τ_s increases, the number of atoms which reside at the surface increases as well. A situation might occur in which the surface atoms determine the behavior of the maser and in which the volume can be treated as a perturbation in much the same way as the surface is treated as a perturbation to the volume frequency. Given the above-mentioned various possible applications for such a surface cryogenic hydrogen maser, it is interesting to investigate under what conditions it might operate.

Based on the knowledge of the preceding section, it seems logical to follow a similar approach but now for small τ_v. Eliminating in this case the rapid time dependence $\exp(-i\omega_s t)$, introducing the reduced time $\bar{t} = t/\tau_v$, and transforming to normalized magnetizations and population inversions as previously, leads to a system of coupled equations for the time derivatives of $\bar{B}$, $\bar{M}^i$, and $\bar{\Delta}^i$. Note that due to the increased recombination at the surface the ratio of the number of atoms at the surface to the number of atoms in the volume is no longer given by the relation $N_s/N_v = \tau_s/\tau_v$. Instead, the two are related via

$$\frac{n_v}{\tau_v} = \frac{n_s}{\tau_s} + \gamma_{\rm rec}^s n_s^2 \ , \quad (8)$$

where n_s is the density of atoms per unit volume at the surface (defined as $n_s = \sigma A_b/V_b$) and $\gamma_{\rm rec}^s n_s$ is the recombination rate. Consistent with Sec. II, we have assumed $\dot{n}_v = \dot{n}_s = 0$, which implies a restriction to steady oscillation taking into account that $\gamma_{\rm rec}^s$ depends on the populations of the a and c levels, i.e., on the population inver-

sion, which may be time dependent. An example of the behavior of n_s and n_v as a function of temperature is displayed in Fig. 2 for a flux of 2×10^{13} atoms per second and an area-to-volume ratio of 300 cm^{-1}. At temperatures below 500 mK the temperature dependence of the densities starts to deviate from the curves obtained by neglecting the recombination. This is due to the fact that a significant part of the incoming flux is needed to compensate for recombination at the surface, instead of building up the density. As, however, the recombination time is still large compared to the residence time at the surface, the equality $n_s/n_v = \tau_s/\tau_v$ is still valid. Only for temperatures below 80 mK, when the volume density starts to increase again, is the recombination time equal to or smaller than the residence time and is Eq. (8) needed to describe the relation between n_s and n_v.

In the spirit of the preceding section, we now try to express $\overline{M}^v$ and $\overline{\Delta}^v$ in the corresponding surface quantities. This is possible by neglecting all terms containing τ_v in the dynamical equations for $\overline{M}^v$ and $\overline{\Delta}^v$. It is essential, however, to keep the τ_v terms containing $\overline{M}^s, \phi_1 = \tau_v(\omega_v - \omega_s)$, Δ_0^v, and $\overline{\Delta}^s$. Here we take into account that ϕ_1 is large compared to 1, that $\overline{M}^s$ and $\overline{\Delta}^s$ have to be included to retain the coupling between surface and volume, and that the Δ_0^v term is essential for maintaining the population inversion. The result is

$$\overline{M}^v = \frac{1}{1+i\phi_1} \frac{\tau_v N_s}{\tau_s N_v} \overline{M}^s ,$$

$$\overline{\Delta}^v = \frac{\tau_v N_s}{\tau_s N_v} \overline{\Delta}^s + \tau_v \gamma_\parallel^v \frac{\Delta_0^v}{N_v} . \tag{9}$$

Note that M^v is 90° out of phase with M^s. Substituting these expressions in the remaining equations and including $O(\tau_v)$ terms again leads to the usual maser equations (1) with the effective parameters

$$\omega_{\text{at}} = \omega_s + \frac{1}{\tau_s} \frac{\phi_1}{1+\phi_1^2} ,$$

$$\gamma_\perp = \gamma_\perp^s + \frac{1}{\tau_s} \frac{\phi_1^2}{1+\phi_1^2} , \tag{10}$$

$$\Delta_0 = \Delta_0^s + \frac{\gamma_\parallel^v}{\gamma_\parallel^s} \Delta_0^v .$$

It is interesting to point out that the maser will oscillate at a frequency determined by the hyperfine frequency at the surface, i.e., ω_{at} is very close to ω_s, as indicated in Fig. 3, which should be a characteristic feature of a surface maser. This follows also from our simulations. It is thus indeed possible to make the volume shift small by decreasing the temperature. This means that one of the device-dependent processes that affect the transition frequency can possibly be eliminated. In order to assess the feasibility of the SCHM as a primary time standard, however, other processes which influence the transition frequency, such as collisions, also have to be examined. Another important fact is that $\gamma_\perp$ ($\simeq \gamma_\perp^s + 1/\tau_s$) turns out to have approximately equal contributions from $\gamma_\perp^s$ and dephasing in the volume under the specific conditions of the SCHM that we will examine. Note, furthermore, that the effective value of Δ_0, which deviates significantly from the pure surface value Δ_0^s, is a result of the present treatment and does not follow from the method of Anderson and Weiss. An analogous significant surface correction
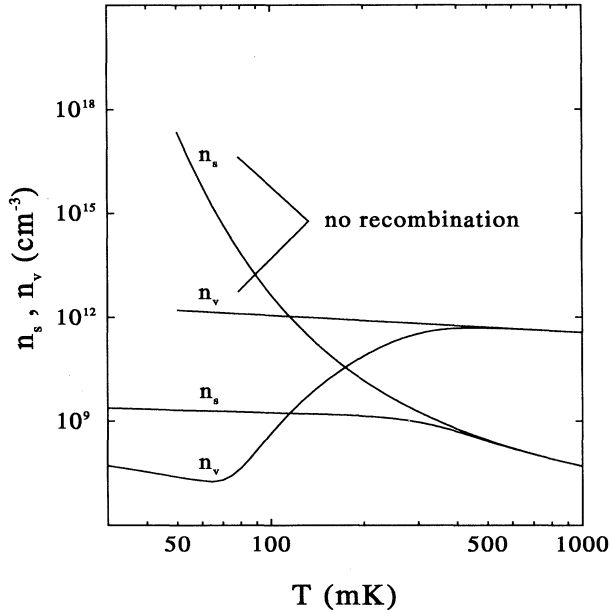

FIG. 2. Surface density per unit volume n_s and volume density n_v as a function of temperature for a flux of 2×10^{13} atoms per second and an area-to-volume ratio of 300 cm^{-1}. For comparison the curves corresponding to $n_s/n_v = \tau_s/\tau_v$, i.e., neglecting recombination, are given. The low-temperature part of these curves, where the surface density saturates, is omitted.
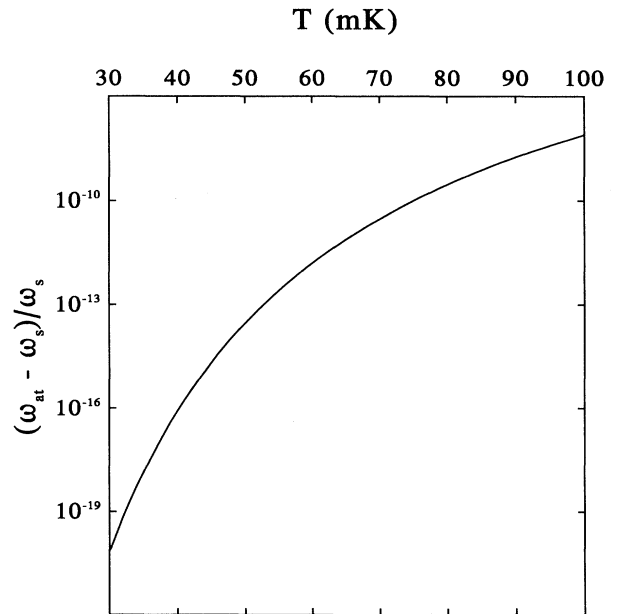

FIG. 3. Ratio of the volume shift as given by Eqs. (10) to the transition frequency as a function of temperature for an area-to-volume ratio of 300 cm^{-1}.

to Δ_0^v did not occur in the previous case of the CHM.

In order to assess the realizability of a surface maser these parameters have to be substituted in the oscillation condition, which on resonance is given by

$$g^2\Delta_0 > \kappa\gamma_\perp .\qquad(11)$$

For usual values of the incident flux the minimum required cavity Q value is determined by $\gamma_\perp$, i.e., by the surface density and the temperature via $\gamma_\perp^s$ and τ_s. For typical device parameters we have taken the values common to the current hydrogen maser, as mentioned in Sec. II, with the exception of the area-to-volume ratio of the storage bulb, where we used the value given in Ref. [7]. For the temperature-density combination of 70 mK and 10^9 cm^{-3} we then find the oscillation condition to be satisfied with $Q_c = 3 \times 10^6$. This result is confirmed by a numerical simulation of the unapproximated set of Eqs. (5), leading to Figs. 4–9. Both for higher temperatures and smaller area-to-volume ratios the oscillation condition requires higher Q_c values. In Figs. 4–6 the output power of the maser is shown as a function of density, temperature, and cavity quality factor. Figure 4 shows the importance of recombination to obtain maser oscillation: instead of the usual characteristic quadratic dependence of the power on the density with a decrease at higher densities, the power now continues to increase. In the CHM the influx of atoms is proportional to the density. At low densities Δ_0 is determined by this influx whose increase is sufficient to compensate for the accompanying increase of $\gamma_\perp$. At high densities, however, the increase of Δ_0 with density is less than linear due to longitudinal relaxation, eventually making oscillation impossible. Due to recombination the influx is a quadratic function
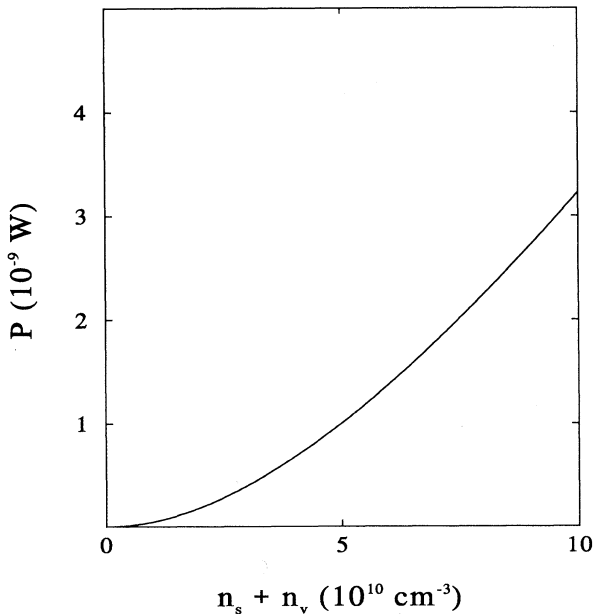
FIG. 5. The output power of the a maser as a function of temperature for $A_b/V_b = 300$ cm^{-1}, $n_s + n_v = 10^9$ cm^{-3}, $Q_c = 3 \times 10^6$ and 3×10^7, and $\omega_c = \omega_s$.

of the density in the SCHM. In this case the oscillation condition will always be fulfilled above a certain minimum density. The sharp decrease of the power for higher temperatures displayed in Fig. 5 indicates the importance of the dephasing in the volume on the effective atomic linewidth. At the low-temperature side $\gamma_\perp$ is determined by $\gamma_\perp^s$. At the high-temperature side the de-

FIG. 4. The output power of the a-c maser as a function of the total density in the storage bulb for $A_b/V_b = 300$ cm^{-1}, $T = 70$ mK, $Q_c = 3 \times 10^6$, and $\omega_c = \omega_s$.
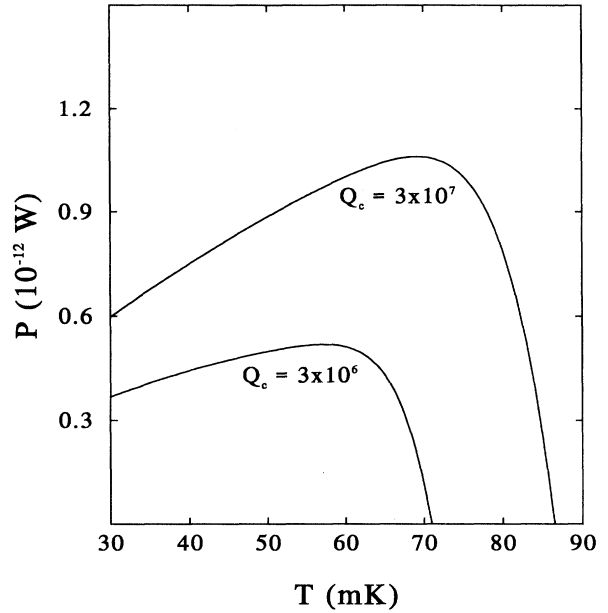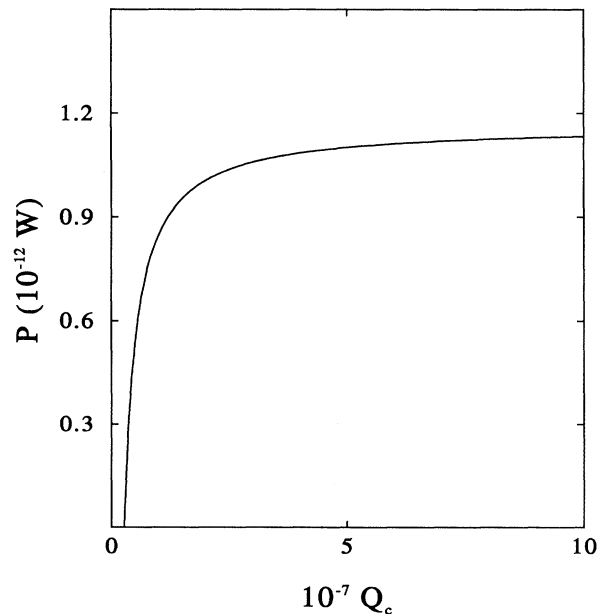
FIG. 6. The output power of the a-c maser as a function of the cavity quality factor for $A_b/V_b = 300$ cm^{-1}, $n_s + n_v = 10^9$ cm^{-3}, $T = 70$ mK, and $\omega_c = \omega_s$.
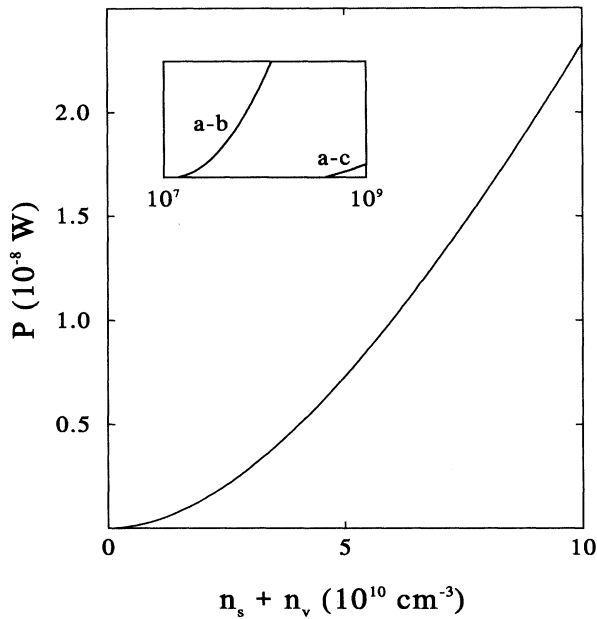
FIG. 7. Same as Fig. 4 but for the a-b maser. The inset shows the minimum required densities for maser oscillation for both the a-b and a-c maser obtained by magnifying the left-hand sides of Fig. 4 and this figure.

phasing term becomes more important. This results in a rapid increase of the transverse relaxation rate which inhibits maser oscillation. The relationship between the power and the cavity quality factor in Fig. 6, finally, implies the expected linear relationship between the number of photons in the cavity and the quality factor ($|B|^2 \sim PQ_c$).

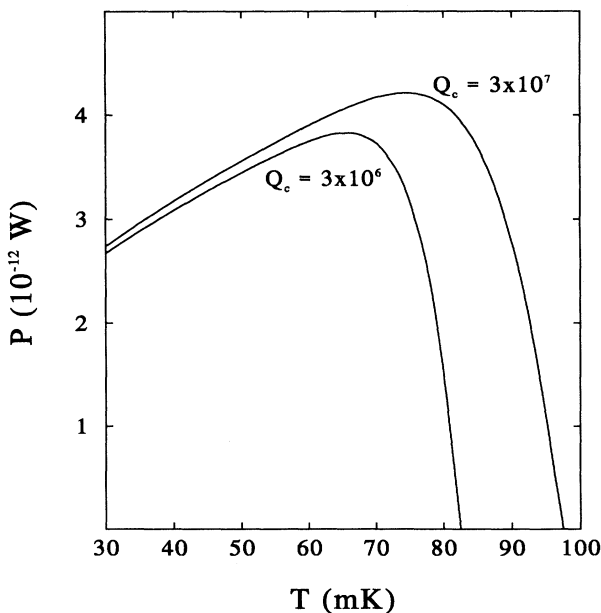To shed further light on the interesting role of recom-

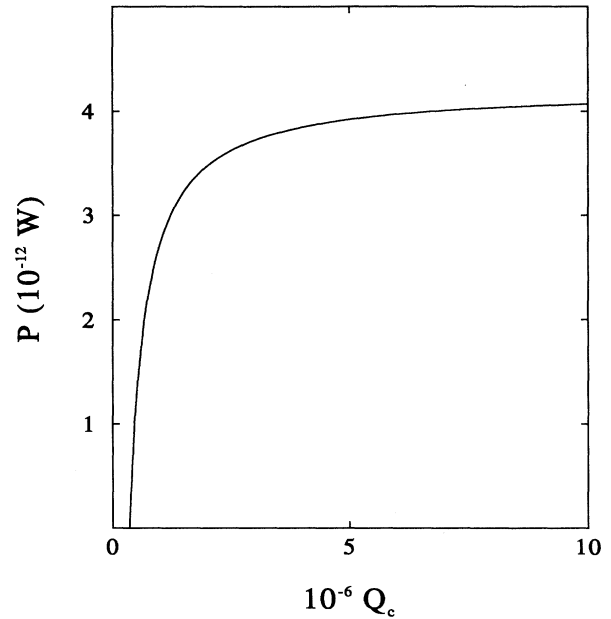FIG. 8. Same as Fig. 5 but for the a-b maser.

FIG. 9. Same as Fig. 6 but for the a-b maser.

bination it is instructive to see what happens if surface recombination is absent. Without recombination the transverse relaxation rate as given by Eqs. (10) will be greatly reduced, which might suggest a favorable condition for operation of a surface maser. An evaluation of the oscillation condition (11), however, shows that in this case no oscillation can exist for reasonable maser parameters. The main cause is the importance of Δ_0^v in sustaining maser oscillation. In order for the maser to operate, enough population-inverted influx should be supplied to the storage bulb per unit time. In the absence of recombination, however, the flux of new atoms into the storage bulb equals the flux out. This flux is determined by the number of atoms in the volume, which is only a small fraction of the total number of atoms in the storage bulb under the surface-maser conditions examined above (see Fig. 2). The flux of new population inversion $\gamma_\parallel^v \Delta_0^v$ is then too small to support maser oscillation. With recombination the incoming flux not only compensates the outgoing flux, but in order to maintain a constant density the atoms which disappear due to recombination must also be replaced. This implies a larger influx of new population inversion for the same value of the surface density and thus a more favorable condition for maser oscillation.

In the previous considerations we needed Δ_0^v for oscillation. A fascinating alternative would be a surface maser based on the a-b transition. In this case, due to preferential recombination and relaxation of the a atoms, Δ_0^s is positive, i.e., oscillation can in principle be realized even without population inversion in the incoming beam. However, it is possible to feed the storage bulb with an incoming beam of b atoms (this will be discussed in the next section). The same analysis which has been applied to the a-c maser can now be used for the a-b maser. For the same operating conditions as above both the oscilla-

tion condition and the numerical simulations show that oscillation is already possible for $Q_c = 4 \times 10^5$, which is almost 10 times lower than for the a-c maser. More advantages are clearly shown in Figs. 7–9. Not only does the a-b maser operate at much lower Q_c values, for equal Q_c values it also operates at lower densities and, even more important, at higher temperatures. These higher temperatures could possibly make the a-b maser easier to operate.

Comparing the conditions under which the SCHM operates with the normal operating conditions of the CHM it is clear that for the former a cavity quality factor is needed which is several orders of magnitude larger. Partially, this is due to the magnitude of Δ_0, which involves different physics. In the CHM the influx of atoms into the storage bulb replaces directly the atoms flowing out. In the SCHM new atoms primarily replace atoms which have recombined. The increase of Q_c is also due to the larger transverse relaxation rate. In the CHM both the storage time of the atoms and the collisional relaxation time are of the order of one second with only a small contribution from dephasing at the surface. For our example temperature-density combination of 70 mK and 10^9 cm^{-3}, in contrast, the resonant linewidth $\gamma_\perp$ is equal to about 50 s^{-1} with approximately equal contributions from dephasing in the volume and $\gamma_\perp^s$. To compensate for this rapid loss of coherence, a much better cavity is needed.

V. EXPERIMENTAL CONSIDERATIONS

In this section we discuss some experimental aspects connected with the realization of a SCHM. We first consider some cryogenic questions. Temperatures of the order of 50–100 mK are easily attainable with a ^{3}He-^{4}He dilution refrigerator, with cooling powers of a few hundred microwatts. Recombination of hydrogen can put a large heat load on the refrigerator. We calculate the heat load for the SCHM we have analyzed, with $\sigma = n_s V_b / A_b = 3 \times 10^6$ cm^{-2} where the area $A_b = 6 \times 10^4$ cm^2. The heat produced is

$$\dot{Q} = K_s \sigma^2 A_b \frac{D_0}{2} , \qquad (12)$$

where $D_0 = 7 \times 10^{-19}$ J is the recombination energy released per pair of atoms. Using the experimental value of the recombination constant $K_s = (3 \times 10^{-5}) \mathrm{K}^{-1/2} \sqrt{T}$ cm^2 s^{-1} from Morrow et al. [9] we find 2 μW, which is easily absorbed by a dilution refrigerator. A connected problem posed by temperatures in the range 50–100 mK is that a gas of hydrogen loses thermal contact due to the Kapitza resistance between the helium film and the substrate. However, in the maser described here, due to the low hydrogen-surface densities, the heat production is substantially lower than has been experimentally studied in the past. Thus the thermal gradients which develop may be smaller, resulting in some flexibility in the range of accessible temperatures.

Finally, we consider the filling fluxes required for the SCHM. These are of the order of 10^{13} atoms per second. Such fluxes are easily achieved with existing discharge
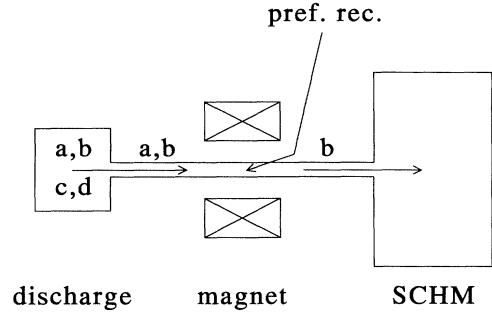

FIG. 10. Schematic picture of an experimental design to load pure b-state atoms in the SCHM.

sources which can deliver 10^{14}–10^{16} atoms per second to a cell.

One of the interesting ideas to emerge from this study is the possibility of a maser working on the a-b transition. An experimental design which can produce a flux of almost pure b-state atoms is shown in Fig. 10. A low-temperature discharge produces hydrogen atoms populating the four hyperfine states. A small (~ 1–2-T) magnet repels the c and d atoms and confines the a and b atoms in a volume at a low temperature (~ 300 mK). This density of atoms converts to pure b-state atoms by preferential recombination. The magnetic field and temperature of this volume can be adjusted so that b-state atoms can reach the maser by thermal escape with a flux sufficient for the a-b SCHM. A small superimposed field can prevent Majorana transitions among the hyperfine states.

VI. DISCUSSION AND CONCLUSIONS

The operation of the sub-Kelvin hydrogen maser is usually described by the Maxwell-Bloch equations. Short periods during which the atoms stick to the surface enter these equations through effective values for the atomic transition frequency and transverse relaxation rate. We have extended these Maxwell-Bloch equations to a description in which the surface is treated symmetrically with the volume, rather than as a perturbation, opening in particular the possibility to treat the pulsed operation of the maser in a regime where the sticking time is comparable to the pulse duration. Work along this line is under way [13]. We have shown that for current versions of the sub-Kelvin hydrogen maser, i.e., for short sticking times, the usual equations and effective parameters are recovered.

Starting from this alternative description, we have investigated the possibilities of constructing a maser whose properties are mainly determined by atoms which reside at the surface, i.e., a surface maser. We have shown that for a maser which is based on the traditional scheme of a transition between the a and c levels of the hydrogen ground state a surface maser is realizable for realistic experimental parameters, for instance, $T = 70$ mK, $n_s + n_v = 10^9$ cm^{-3}, $A_b / V_b = 300$ cm^{-1}, $Q_c = 3 \times 10^6$. This result was obtained both by a simulation of the full set of Eqs. (5) and on the basis of the effective surface

equations. The prospects for an a-b maser are even more favorable.

Apart from the intrinsic interest in the operation of the SCHM, such as the interesting role of recombination and the possibility to observe pulsed oscillation, two important motives for studying the influence of the surface on the maser operation and in particular the SCHM are the possibility of creating a primary time standard and the precision measurement of properties connected with a two-dimensional gas of atomic hydrogen.

In this connection one could think of an extension of the measurement of the surface adsorption energy in Ref. [11] to a situation with lower surface densities, made possible by the greater sensitivity, with the associated advantage of a more reliable temperature determination. Another possibility worth considering would be to measure $\gamma_\parallel^s$ and $\gamma_\perp^s$ as a function of surface density and field orientation. In principle, this would enable an independent determination of the dipolar T_2 [11]. This could stimulate the development of a satisfactory theory of line broadening including the coherent contribution of all dipolar fields. The main parts of $\gamma_\parallel^s$ and $\gamma_\perp^s$, due to recombination, would also be a welcome addition to the knowledge desired for developing a satisfactory theory of surface recombination. Apart from the line broadening, much interest has been devoted in the past to the effect of dipolar and spin-exchange collisions on the transition frequency itself. The sensitivity of the maser could allow a very accurate determination of the collisional shift parameters, especially since the output is not directly disturbed by a bulk signal.

In order to obtain a primary time standard it is essential that the effective atomic frequency ω_{at} be independent of specific device parameters, such as those which determine the wall (volume) shift, but also spin-exchange and dipolar shifts, the influence of the substrate, and the thickness of the helium film. From our analysis it follows that the volume shift can be suppressed in the SCHM. It is very unlikely, however, that the influence of the other processes can also be sufficiently reduced. For example, Morrow and Berlinsky [10] have calculated the spin-exchange frequency shift for hydrogen atoms adsorbed on a surface. Although in this calculation the motion of the atoms perpendicular to the surface is neglected it gives a good indication whether this effect seriously influences the operation of the SCHM. For our reference temperature-density combination this leads to a frequency shift $\Delta\omega_{at} \simeq 10$ rad s^{-1}, given the fact that $(\rho_{cc} - \rho_{aa})$ is approximately 0.1 in our simulations. It is clear that already this effect is much too large to open possibilities for the SCHM as a primary time standard.

Under the conditions studied the resonance linewidth $\gamma_\perp$ ($\simeq 50$ s^{-1}) is still much smaller than the typical cavity loss rate κ ($\simeq 1500$ s^{-1}). Although the ratio between these two is less than in the CHM, fluctuations in the cavity frequency are still sufficiently suppressed to measure with great accuracy the above-mentioned surface processes.

ACKNOWLEDGMENTS

[1] S. B. Crampton, W. D. Phillips, and D. Kleppner, Bull. Am. Phys. Soc. **23**, 86 (1978); R. F. C. Vessot, M. W. Levine, and E. M. Mattison, NASA Technical Memorandum No. 78104, 1978 (unpublished), p. 549; A. J. Berlinsky and W. N. Hardy, NASA Conference Publication No. 2220, 1982 (unpublished), p. 547.

[2] H. F. Hess, G. P. Kochanski, J. M. Doyle, T. J. Greytak, and D. Kleppner, Phys. Rev. A **34**, 1602 (1986); M. D. Hürlimann, W. N. Hardy, A. J. Berlinsky, and R. W. Cline, ibid. **34**, 1605 (1986); R. L. Walsworth, I. F. Silvera, H. P. Godfried, C. C. Agosta, R. F. C. Vessot, and E. M. Mattison, ibid. **34**, 2550 (1986).

[3] A. C. Maan, H. T. C. Stoof, B. J. Verhaar, and P. Mandel, Phys. Rev. Lett. **64**, 2630 (1990); **65**, 2319 (1990); P. Mandel, A. C. Maan, B. J. Verhaar, and H. T. C. Stoof, Phys. Rev. A **44**, 608 (1991).

[4] W. N. Hardy, M. D. Hürlimann, and R. W. Cline, Jpn. J. Appl. Phys. **26**, 2065 (1987); M. D. Hürlimann, Ph. D. thesis, University of British Columbia, Canada, 1989 (unpublished).

[5] P. W. Anderson and P. R. Weiss, Rev. Mod. Phys. **25**, 269 (1953); P. W. Anderson, J. Phys. Soc. Jpn. **9**, 316 (1954); A. Abragam, The Principles of Nuclear Magnetism (Claren-don, Oxford, 1961).

[6] M. Morrow and W. N. Hardy, Can. J. Phys. **61**, 956 (1983).

[7] L. Pollack, S. Buchman, and T. J. Greytak, Phys. Rev. B **45**, 2993 (1992).

[8] B. J. Verhaar, J. M. V. A. Koelman, H. T. C. Stoof, O. J. Luiten, and S. B. Crampton, Phys. Rev. A **35**, 3825 (1987); J. M. V. A. Koelman, S. B. Crampton, H. T. C. Stoof, O. J. Luiten, and B. J. Verhaar, ibid. **38**, 3535 (1988).

[9] M. Morrow, R. Jochemsen, A. J. Berlinsky, and W. N. Hardy, Phys. Rev. Lett. **46**, 195 (1981); **47**, 455 (1981); D. A. Bell, H. F. Hess, G. P. Kochanski, S. Buchman, L. Pollack, Y. M. Xiao, D. Kleppner, and T. J. Greytak, Phys. Rev. B **34**, 7670 (1986).

[10] M. Morrow and A. J. Berlinsky, Can. J. Phys. **61**, 1042 (1983).

[11] I. Shinkoda and W. N. Hardy, J. Low Temp. Phys. **85**, 99 (1991).

[12] A. Nayfeh, Perturbation Methods (Wiley, New York, 1973).

[13] P. Mandel (private communication).