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MODIFIED ZEEMAN HYPERFINE SPECTRA OBSERVED IN H^1 AND Rb^{87} GROUND STATES INTERACTING WITH A NONRESONANT rf FIELD

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We have observed new effects in the Zeeman hyperfine spectra of H^1 and Rb^{87} when a nonresonant linear radiofrequency field is applied perpendicular to the static field: The Zeeman lines coalesce in some cases; new sideband resonances also appear. A theoretical explanation is given for these effects. Some possible applications for atomic clocks and masers are considered.

The Zeeman hyperfine spectrum of an atomic ground state is drastically modified when a linear radiofrequency field, $\vec{H}_1 \cos\omega t$, is applied perpendicular to the static field $\vec{H}_0$: A decrease in the splitting between the Zeeman lines, which in some cases coalesce, has been observed; new sideband resonances have also been detected. The rf field which produces these effects is nonresonant either for the Zeeman or for the hyperfine transitions: ω is small compared with the hyperfine separation, but much larger than the Larmor precession frequency $\omega_0 = \gamma H_0$ (γ is the gyromagnetic ratio of the ground-state levels). We have studied these phenomena in optically pumped Rb^{87} atoms and state-selected H^1 atoms in a hydrogen maser.

The Rb^{87} experimental setup is a classical one [Fig. 1(a)]. The sample is a 3-cm-diam paraffin-coated cell without buffer gas; it is protected from magnetic noise by a large five-layer cylindrical Mumetal shield.[1] The Rb^{87} atoms are optically pumped by the D_2 component of a nonpolarized light beam B emitted by a Rb^{87} lamp and passing through a Rb^{85} filter in order to achieve hyperfine pumping.[2] The microwave field, $\vec{h}_1 \cos\Omega t$, is delivered by a horn and comes, through several stages of frequency multiplication, from a 5-MHz quartz crystal oscillator; $\vec{h}_1$ is perpendicular to the static field $\vec{H}_0$. The frequency $\Omega/2\pi$ can be swept over a range of a few kHz around the frequency $\Omega_0/2\pi$ of the $(F = 2,\ M_F = 0) \leftarrow (F = 1,\ M_F = 0)$ field-independent transition ($\Omega_0/2\pi = 6834.683$ MHz). The variation of the hyperfine-level populations is monitored by the intensity of the transmitted light measured by a photomultiplier. Due to the polarization of $\vec{h}_1$, only the transitions $\Delta m_F = \pm 1$ [arrows on Fig. 1(b)] are induced. The two Landé factors of the $F = 1$ and $F = 2$ levels are of opposite sign but have the same absolute value, so that only four equidistant resonances can be observed on Fig. 1(c) (β_1 and β_2 have the same frequency, as do γ_1 and

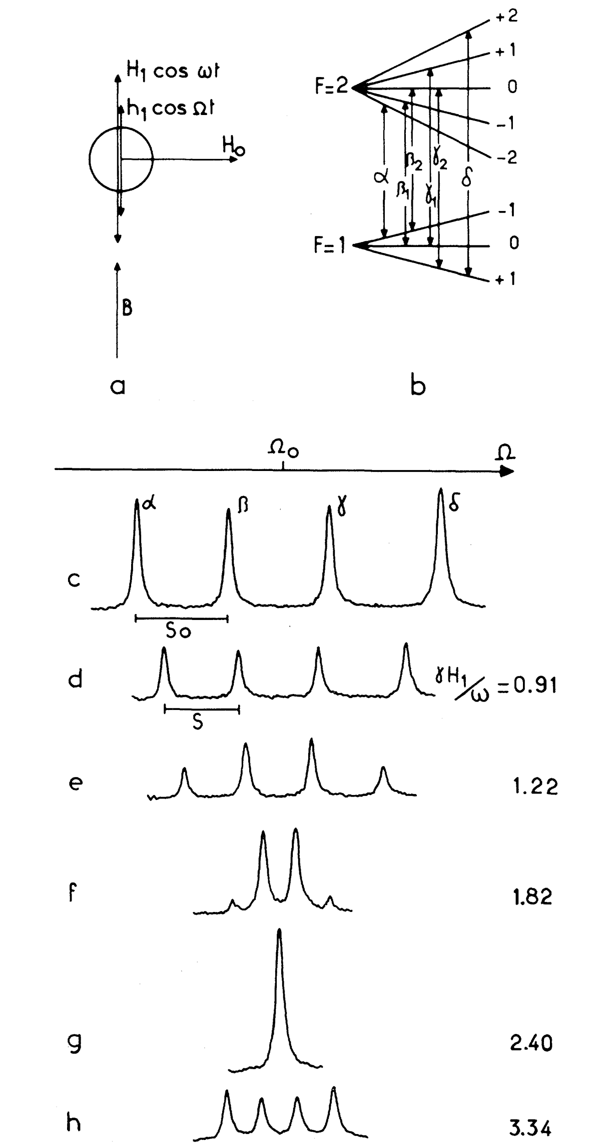

FIG. 1. (a) Schematic diagram of the Rb^{87} experimental setup. (b) Hyperfine Zeeman diagram of the Rb^{87} ground state showing the $\Delta m_F = \pm 1$ transitions. Note the β_1-β_2 and γ_1-γ_2 coincidences. (c) Recording of the $\Delta m_F = \pm 1$ Rb^{87} hyperfine spectrum in a field $H_0 = 0.71$ mG ($s_0/2\pi = 2\omega_0/2\pi = 1000$ Hz). The frequency Ω is swept by tuning the 5-MHz quartz crystal oscillator. The microwave field is square modulated at 2 Hz and phase-sensitive detection is used. (d)-(h) Recordings of the Rb^{87} hyperfine spectrum in the same H_0 field for increasing values of the amplitude of the rf field $H_1 \times \cos\omega t$ measured by the dimensionless quantity $\gamma H_1/\omega$ ($\omega/2\pi = 2700$ Hz).

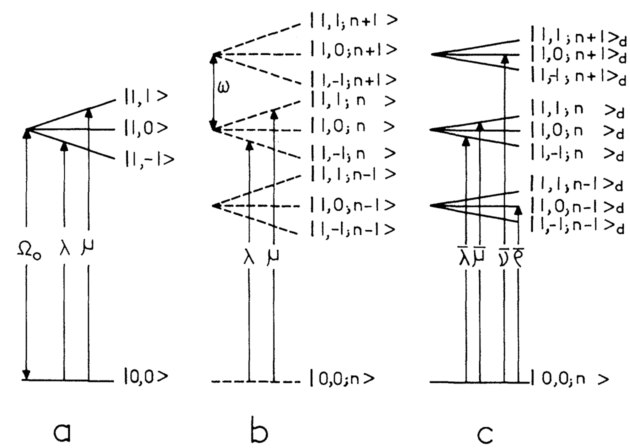

FIG. 2. (a) Hyperfine Zeeman diagram of the H ground state. (b) Energy levels $|F, m_F; n\rangle$ of the "ground state + rf field" system, neglecting the coupling with the rf field. (c) Energy levels $|F, m_F; n\rangle_d$ of the "dressed" hydrogen ground state. Note the change in the slope of the energy levels.

γ_2), the separation between two consecutive lines being $s_0 = 2\omega_0$. When an rf field $\vec{H}_1\cos\omega t$ perpendicular to H_0 and parallel to $\vec{h}_1$ is applied, the hyperfine spectrum is strongly modified, as may be seen on Figs. 1(d)-1(h), corresponding to increasing values of H_1. There are always four equidistant resonances, but their splitting s decreases continuously, cancels for a certain value of H_1 [Fig. 1(g)], and then increases (in fact, the sign of s changes as we will see later on).

The same experiment has been done with a hydrogen maser.[3] The hyperfine spectrum of H is shown on Fig. 2(a). A magnetic state selector provides a difference of populations between the hyperfine states $F = 1$, $m_F = 0, 1$ and $F = 0$, $m_F = 0$. The oriented atoms are stored in a Teflon-coated 16-cm-diam bulb placed in a five-layer magnetic shield. The arrangement of the fields $\vec{H}_0$, $\vec{h}_1$, and $\vec{H}_1$ is the same as for the Rb^{87} experiment (the direction of $\vec{h}_1$ is determined by the cavity mode of the maser). Because of the polarization of $\vec{h}_1$, only the two field-dependent transitions (λ, μ) can be induced. The self-oscillation of the maser which produces the $\vec{h}_1$ field ($\Omega_0/2\pi = 1420.405$ MHz) is observed only in the μ transition (there is no population iversion for the λ transition). We have measured the frequency of oscillation in a given static field $H_0 = 14$ μG as a function of the rf field amplitude H_1 ($\omega/2\pi = 110$ Hz). We have seen, as in the previous experiment, that the separation $\frac{1}{2}s$ between this oscillation frequency and the one corresponding to the field-independent transition is no longer $\omega_0 = \frac{1}{2}s_0$, but becomes smaller and smaller as the rf field amplitude H_1 increases. This separation exhibits the same behavior when H_1 is varied as described in the Rb^{87} case. On Fig. 3, we have plotted for

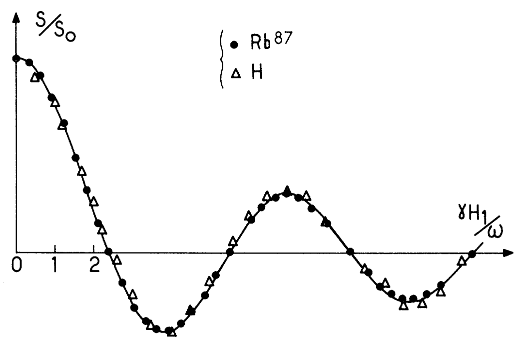

FIG. 3. Plot of the ratio s/s_0 as a function of $\gamma H_1/\omega$. The experimental points for Rb^{87} and H fit into the same theoretical curve.

both experiments the ratio s/s_0 as a function of the dimensionless quantity $\gamma H_1/\omega$, proportional to the rf field amplitude. It can be seen that the experimental results for H^1 and Rb^{87} fit into the same curve.

These results can be understood if one considers that the microwave field $\vec{h}_1 \cos\Omega t$ is a probe which explores the energy diagram of the compound system "atom + rf field" which we call the atom "dressed" by the rf photons. We have already studied in great detail the effect of such a "dressing" on the magnetic properties of an atomic level.[4,5] Let us recall briefly the results of the theory in the simple case of hydrogen. The energy diagram of the free-hydrogen ground state in the field H_0 is given on Fig. 2(a). In the presence of an rf field $\vec{H}_1 \cos\omega t$ perpendicular to $\vec{H}_0$, these energy levels are modified. First, suppose that H_1 is very small so that the coupling between the atomic system and the rf photons can be neglected. Then the energy levels of the compound system will merely be the states $|F, m_F; n\rangle$ representing the atom in the state $|F, m_F\rangle$ $(F=1, 0)$ with n rf photons present; the energy of these states is (with $\hbar=1$) $n\omega$ if $F=0$, and $\Omega_0 + m_F\omega_0 + n\omega$ if $F=1$. In the $F=1$ states, the energy diagram of the compound system will consist of manifolds separated from each other by the energy ω; each manifold corresponds to a given value of n and is split into three magnetic levels corresponding to the three possible m_F values [dashed lines on Fig. 2(b)]. A microwave field can induce only $\Delta F=1$, $\Delta n=0$ transitions [for example when $\vec{h}_1$ is perpendicular to $\vec{H}_0$, only the transitions λ and μ of Fig. 2(b) are possible]. The selection rule $\Delta n=0$ results from the commutation of microwave and rf variables. The coupling with the rf field which we now take into account occurs only in the $F=1$ states and leads to a kind of "renormalization" of the "unperturbed" system described above. It has two effects[5]: First, it changes the slope of the energy levels [full lines on Fig. 2(c)]; this corresponds to a modification of the Landé factor g_F of the hyperfine level F, which becomes now

$$\bar{g}_F = g_F J_0(\gamma_F H_1/\omega), \quad \gamma_F = g_F \mu_B, \tag{1}$$

where J_0 is the zero-order Bessel function and μ_B the Bohr magneton. Second, the coupling modifies the energy eigenstates: The "renormalized" states $|F, m_F; n\rangle_d$ are now admixtures of the unperturbed states $|F, m_F'; n'\rangle$ due to virtual absorptions and emissions of rf quanta and no longer correspond to a definite n value.

The modification of the Landé factor explains our experimental observations. In the H-maser experiment, we detect the maser oscillation on the transition $\bar{\mu}$ $(|F=0; n\rangle \to |F=1, m_F=+1; n\rangle_d)$ [Fig. 2(c)] of the "dressed" atom which corresponds, for $H_1=0$, to the field-dependent transition μ of the free atom [Fig. 2(a)]. The case of Rb^{87} is more complicated because both hyperfine levels $F=2$, $F'=1$ are coupled to the rf field. But relation (1) holds for both hyperfine levels and since $\gamma_F = -\gamma_{F'}$, and J_0 is an even function, $\bar{g}_F$ and $\bar{g}_{F'}$ are modified in the same way and in particular cancel for the same values of H_1. For this reason, the splitting s between the field-dependent resonances must vary exactly as in the hydrogen case. On Fig. 3 we have plotted in solid lines the theoretical curve $J_0(\gamma_F H_1/\omega)$ which fits very well with the experimental points. We have observed several oscillations of s. Let us mention that the variations of $\bar{g}_F$ are responsible for other physical effects such as the modification of the width of the zero-field level-crossing resonances (Hanle effect).[6]

As can be seen on Fig. 1 in the case of Rb^{87}, the coupling with the rf field affects not only the splitting s but also the intensity of the lines. This is due to the modification of the magnetic dipole matrix elements between the corresponding perturbed eigenstates. Moreover, new transitions can now be induced between two eigenstates $|F, m_F; n\rangle_d$ and $|F', m_{F'}'; n'\rangle_d$ with different n values (as n is no longer a good quantum number, the selection rule $\Delta n=0$ is no longer valid). Thus, new sideband resonances at the frequencies

$$\Omega = \Omega_0 + (n-n')\omega + (\bar{g}_F m_F - \bar{g}_{F'} m_{F'}')\mu_B H_0 \tag{2}$$

must appear. They can be understood in terms of simultaneous absorption of one microwave and $n-n'$ rf photons. The position of the observable lines on each sideband may thus be deduced from angular momentum conservation requirements during these absorption processes and obey the following rule, valid for hydrogen as well as for Rb^{87} ($\vec{H}_1$ being always perpendicular to $\vec{H}_0$): If $\vec{h}_1$ is perpendicular to $\vec{H}_0$, the only observable lines correspond to $n-n'$ and $\Delta m_F = m_F - m_{F'}'$ having opposite parities in relation (2) (if $n-n'$ is even, Δm_F is odd, and reciprocally). If $\vec{h}_1$ is parallel to $\vec{H}_0$, $n-n'$ and Δm_F must have the same parity.[7] We have verified these selection rules for Rb^{87} when $\vec{h}_1$ is perpendicular to $\vec{H}_0$: For $|n-n'| = 0$ and 2, the Zeeman pattern consists of four Δm_F-odd lines as on Fig. 1; for $|n-n'| = 1$, we have observed only three Δm_F-even lines (with the same separation s as in the previous case). In the case of hydrogen, when $\vec{h}_1$ and $\vec{H}_1$ are parallel, it turns out that the intensity of all the side bands ($n-n' \neq 0$) vanishes[5] so that only the central resonances ($n-n' = 0$) can be observed. We have verified this point. The side bands could however be observed if $\vec{h}_1$ and $\vec{H}_1$ had two different directions in the plane perpendicular to $\vec{H}_0$. For example, on the first side bands $|n-n'| = 1$; only the two field-independent $\Delta m_F = 0$ transitions $\bar{\nu}$ and $\bar{\rho}$ should appear [see Fig. 2(c)].

A few applications of the previous effects might be considered. For instance, by canceling the Landé factor in both hyperfine levels in alkali metals, one can make all the lines contribute to the field-independent transition and thus increase its intensity. This would be useful for atomic clocks, especially if no efficient pumping between the two $m_F = 0$ states is available. The effect of the magnetic field inhomogeneities on the atomic system might also be reduced by canceling the Landé factor of the "dressed atom." One could also make a maser oscillate on a sideband, field-independent transition; this would provide (by varying ω) a simple way for sweeping a field-insensitive oscillation frequency.

[1]For a description of the magnetic shield, see J. Dupont-Roc, S. Haroche, and C. Cohen-Tannoudji, Phys. Letters 28A, 638 (1969).

[2]P. L. Bender, E. C. Beaty, and A. R. Chi, Phys. Rev. Letters 1, 311 (1958).

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[4]C. Cohen-Tannoudji and S. Haroche, Compt. Rend. 262, 37 (1966). See, also, C. Cohen-Tannoudji and S. Haroche, in *Polarisation, Matière et Rayonnement, Alfred Kastler's Jubilee Book*, edited by the French Physical Society (Presses Universitaires de France, Paris, 1969).

[5]C. Cohen-Tannoudji and S. Haroche, J. Phys. (Paris) 30, 153 (1969).

[6]C. Cohen-Tannoudji and S. Haroche, Compt. Rend. 262, 268 (1966).

[7]For a demonstration of these results in the hydrogen case, see Ref. 5.

GIANT ECHOES IN SOLIDS

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The presence of strong rf electromagnetic echoes is reported in silver alloys containing small ($\lesssim$0.1 at.%) amounts of Mn or Ni. The echoes are observable at 4 MHz from 1.8 to 300°K and possess an amplitude directly proportional to the applied magnetic field. The decay constant, T_2, is about 60 μsec and is not strongly dependent on impurity concentration or magnetic field. Echo amplitudes decrease with increasing impurity concentration.

We have observed strong rf electromagnetic echoes from certain silver alloys. The echoes are similar in some respects to nuclear spin echoes[1] and to "fluxoid echoes."[2] They occur when silver metal containing small amounts of transition metals is placed in a magnetic field and excited by a sequence of short rf pulses. It the resonating system is excited by a sequence of two pulses with equal width separated by a time τ, then a primary echo is observed at a time τ and a secondary echo at time 2τ after the second pulse. An oscilloscope picture of representative echo signals is shown in Fig. 1. A weak echo is also observed after a three-pulse sequence[1,3]; the third pulse is applied at time $t = T$ and the echo appears at $t = T + \tau$. Free-induction decays following the individual pulses are not visible, perhaps because of the long (30-40 μsec) recovery time of the apparatus.

The echoes have the following properties: