

# RESONANT TRANSFER OF COHERENCE IN NONZERO MAGNETIC FIELD BETWEEN ATOMIC LEVELS OF DIFFERENT $g$ FACTORS

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Coherence transfer between atomic levels requires the matching of the Larmor frequencies. For levels with different  $g$  factors this occurs only in zero magnetic field. By changing the  $g$  factors through coupling with a nonresonant rf field, one can match the Larmor frequencies and make the transfer possible in nonzero magnetic fields. Resonances in the transfer appear, depending on the rf field amplitude.  $\text{Rb}^{87}\text{-Cs}^{133}$  spin-exchange experiments are reported which provide an illustration of the theory.

Many atomic collision experiments involve an exchange of angular momentum between two atomic states  $A$  and  $B$ : spin- $1$  or metastability- $2$  exchange collisions, various kinds of excitation transfer processes in resonant<sup>3</sup> or nonresonant<sup>4</sup> collisions, and so on. When a static magnetic field  $H_0$  is applied to the interacting atoms, the exchange efficiency becomes quite different for the angular momentum components parallel and perpendicular to  $H_0$ . One can ignore the magnetic field in the exchange process itself which lasts a very short "collision" time,  $\tau_c$ , during which the Larmor precessions around  $H_0$  are negligible; however, between two consecutive collisions, which are separated from each other by a relatively long mean "exchange" time  $T_{AB}$  ( $T_{AB} \gg \tau_c$ ),<sup>5</sup> the transverse (perpendicular to  $H_0$ ) angular momenta of both systems, also called "Zeeman coherences," precess around  $H_0$  at their own Larmor frequencies  $\omega_A = \gamma_A H_0$  and  $\omega_B = \gamma_B H_0$  ( $\gamma_A$  and  $\gamma_B$  are the respective gyromagnetic ratios of  $A$  and  $B$ ). If  $B$  has initially a transverse orientation and if the resonance condition

$$|\omega_A - \omega_B| T_{AB} \lesssim 1 \quad (1)$$

is not fulfilled, the transverse angular momenta transferred from  $B$  to  $A$  in successive collisions are not in phase so that the net amount of coherence exchanged between the two systems is reduced by a factor, increasing with the mismatch frequency  $\omega_A - \omega_B$ , which rapidly becomes very important.<sup>6</sup> In nonzero magnetic field,<sup>7</sup> the exchange of transverse angular momentum between two systems of different  $g$  factors ( $\gamma_A \neq \gamma_B$ ) is considerably less efficient than the transfer of longitudinal (parallel to  $H_0$ ) orientation which remains at rest between two collisions and may thus be easily transferred from one system to the other.

In this paper we show, however, that Zeeman coherence can flow in a resonant way between

two atomic levels in nonzero magnetic field, even if their  $g$  factors are different, provided they are submitted to a nonresonant rf field of convenient direction and amplitude.

Let us consider spin-exchange collisions between two alkali metals of different nuclear spins  $I$ :  $\text{Rb}^{87}$  ( $I = \frac{3}{2}$ ) and  $\text{Cs}^{133}$  ( $I = \frac{7}{2}$ ). Due to the coupling between electronic and nuclear spins, each alkali metal ground state has two hyperfine levels (let us call the upper and lower ones  $F$  and  $F'$ , respectively) of opposite Landé factors ( $g_F = -g_{F'}$ ). The spin-exchange collision itself involves only the electronic spins  $\tilde{S}$  of both alkalis and the nuclear spins are not affected. After the collision, however, the hyperfine interaction  $a\tilde{I} \cdot \tilde{S}$  again couples  $\tilde{I}$  and  $\tilde{S}$  in each atomic species and, as a result, spin exchange couples together the projections  $^F\tilde{S}(\text{Rb})$ ,  $^{F'}\tilde{S}(\text{Rb})$ ,  $^F\tilde{S}(\text{Cs})$ ,  $^{F'}\tilde{S}(\text{Cs})$  of the electronic spin in each hyperfine level of both alkalis.<sup>8</sup> For the sake of simplicity, we shall disregard at first the existence of the lower  $F'$  hyperfine levels and only take into account the coupling between the  $F$  upper levels on each atom. As the two Landé factors are different for the two species [ $g_F(\text{Rb}) = 2g_F(\text{Cs})$ ], the transverse circular components of the spins  $^FS_+(\text{Rb})$  and  $^FS_+(\text{Cs})$  ( $^FS_+ = ^FS_x + i^FS_y$ ) precess at different frequencies so that it seems impossible to transfer Zeeman coherence between  $\text{Rb}^{87}$  and  $\text{Cs}^{133}$ . As is shown however by Haroche *et al.*,<sup>9</sup> the coupling of an atomic state (of total angular momentum  $F$ ) with a nonresonant rf field  $\tilde{H}_1 \cos \omega t$  perpendicular to the static field  $H_0$ , with  $\omega$  large compared with the Larmor frequency, leads to a kind of "dressing" of the atomic system by the rf photons and changes the Landé factor of the free atom,  $g_F$ , which becomes

$$\bar{g}_F = g_F J_0(\gamma_F H_1 / \omega); \quad \gamma_F = g_F \mu_B \quad (2)$$

( $J_0$  is the zero-order Bessel function and  $\mu_B$  the Bohr magneton;  $\hbar = 1$ ). We have plotted as solid

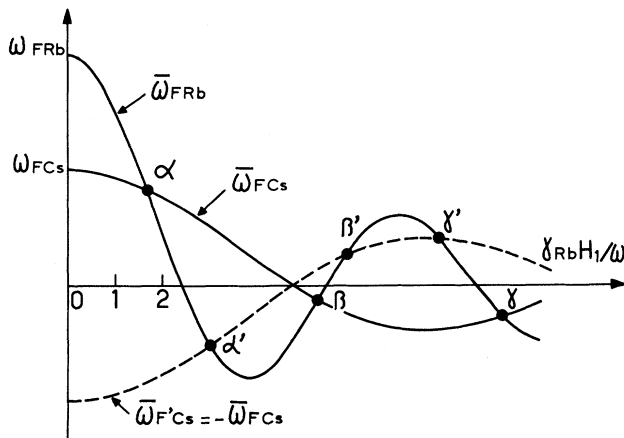


FIG. 1. Plot of  $\text{Rb}^{87}$  and  $\text{Cs}^{133}$  Larmor frequencies in a given static field  $H_0$  as a function of the rf field amplitude measured by the dimensionless quantity  $\gamma_{\text{Rb}} H_1 / \omega$  ( $\gamma_{\text{Rb}}$  is the Rb gyromagnetic ratio). The solid-line curves give the frequencies  $\bar{\omega}_{F\text{Rb}}$  and  $\bar{\omega}_{F\text{Cs}}$  of upper hyperfine levels in both alkalis. The dashed-line curve represents the frequency  $\bar{\omega}_{F'\text{Cs}} = -\bar{\omega}_{F\text{Cs}}$  of the lower  $F'$  level of Cs.

lines in Fig. 1 the corresponding Larmor frequencies in a given static field  $H_0$ ,  $\bar{\omega}_{F\text{Rb}} = \bar{g}_F(\text{Rb}) \times \mu_B H_0$  and  $\bar{\omega}_{F\text{Cs}} = \bar{g}_F(\text{Cs}) \mu_B H_0$  for the  $\text{Rb}^{87}$  and  $\text{Cs}^{133}$  upper hyperfine  $F$  levels, as a function of the rf-field amplitude  $H_1$ . It can be seen that for several values of  $H_1$  (points  $\alpha, \beta, \gamma, \dots$ ), components at the same frequency appear in the preces-

sion of the two "dressed" alkali metal orientations  $^F\bar{S}_+(\text{Rb})$  and  $^F\bar{S}_+(\text{Cs})$ , and therefore exchange of transverse angular momentum becomes possible between the two "dressed" atomic levels.

The experimental setup is shown in Fig. 2(a): A 6 cm diam paraffin-coated cell contains the  $\text{Rb}^{87}$  and  $\text{Cs}^{133}$  atoms. The Cs density is about ten times the Rb density. The sample temperature is regulated to about  $45^\circ\text{C}$ ; at this temperature, the spin-exchange collision processes are predominant over other relaxation mechanisms since the exchange time ( $\sim 0.02$  sec) is short compared with the wall relaxation times of both species ( $\sim 0.2$  sec). A circularly polarized light beam  $B_1$ , produced by a Cs lamp, achieves the orientation of the Cs atoms along the  $0z$  direction of the static field  $H_0$ . By exchange collisions the Rb atoms get oriented in the same direction.

In order to get orientation on Cs only, a saturating rf field tuned to the Rb Larmor frequency is applied perpendicular to  $H_0$ ; this field destroys the longitudinal Rb orientation and is strong enough to introduce no appreciable Rb transverse orientation. At time  $t=0$ , the Rb resonant rf field is removed, the light beam  $B_1$  is switched off by a shutter, and, just after, an rf  $\pi/2$  pulse is applied to the Cs atoms in order to tilt their orientation in the  $0x$  direction [see the sequence of events on Fig. 2(b)]. Thus, just af-

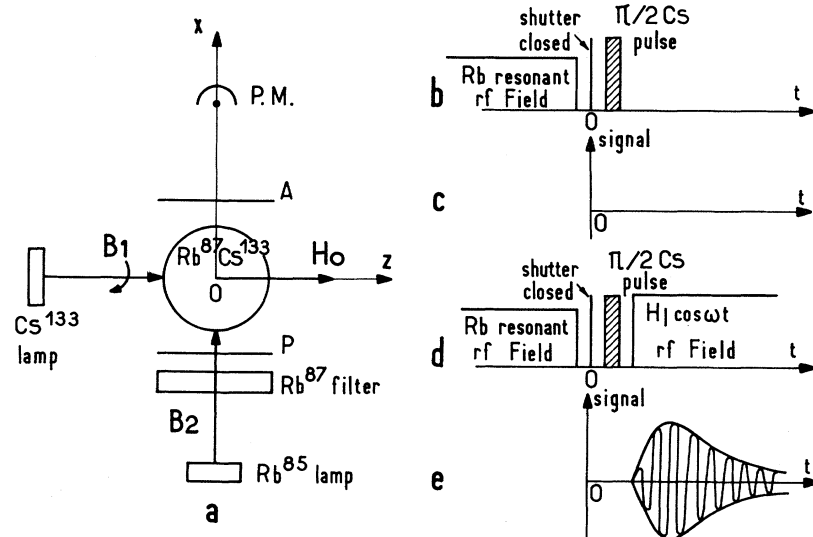


FIG. 2. (a) Schematic diagram of the experimental setup for the  $\text{Rb}^{87}$ - $\text{Cs}^{133}$  spin-exchange experiment. (b) Schematic diagram of the sequence of operations performed in order to introduce transverse angular momentum in Cs. (c) Output of the photomultiplier (P.M.) during and after the sequence of events of (b): There is no coherence transfer at all and no signal is expected to appear. (d) Sequence of operations performed in order to get coherence transfer: One applies just after the  $\pi/2$  Cs pulse an rf "dressing" field which matches the atomic  $g$  factors. (e) Theoretical shape of the transfer ac signal obtained during the sequence of (d). Note that the signal starts just when the rf "dressing" field is applied.

ter this pulse, we have prepared a transverse orientation in the Cs atoms only [ $F S_x(\text{Cs}) \neq 0$ ,  $F S_x(\text{Rb}) = 0$ ]. In order to study how this orientation can leak from Cs to Rb, we use a nonresonant linearly polarized light beam  $B_2$ , produced by a  $\text{Rb}^{85}$  lamp filtered by a  $\text{Rb}^{87}$  cell, which allows the detection by Faraday rotation<sup>10</sup> of the  $\text{Rb}^{87}$  transverse angular momentum  $F S_x(\text{Rb})$  along the  $0x$  direction. The cell is placed between a polarizer  $P$  and an analyzer  $A$  at  $45^\circ$ ; the photomultiplier P.M. monitors the variations of the transmitted light which are proportional to those of  $F S_x(\text{Rb})$ . Note that this nonresonant detection beam does not disturb the Rb spins.

The transverse orientation introduced at time  $t=0$  in Cs will precess around  $H_0$  at the Cs Larmor frequency and decay as a consequence of spin exchange and wall-collision relaxation processes (as the light beam  $B_1$  is switched off, there is no longer optical pumping or optical relaxation). The Larmor precessions being different for both alkali metals, this orientation will not be appreciably transferred to Rb and no signal is expected to appear at the output of the photomultiplier [Fig. 2(c)]. (We have verified this result which checks that there is no Rb coherence introduced during the sequence of operation before time  $t=0$ .) Let us now resume the same experiment, but just after the  $\pi/2$  Cs pulse [see the sequence of operations in Fig. 2(d)], we apply along the  $0x$  direction an rf field  $\tilde{H}_1 \cos \omega t$  whose amplitude  $H_{1\alpha}$  corresponds exactly to the first crossing point  $\alpha$  of the two solid-line Bessel curves of Fig. 1. The Larmor precession frequencies of both systems are now matched at the same value; we observe then at the output of the photomultiplier an ac signal whose frequency,  $\bar{\omega}_{\text{FRb}} = \bar{\omega}_{\text{FCs}}$ , is precisely the Larmor frequency common to both alkali metals at the point  $\alpha$ ; as there was no Rb orientation at all before the rf "dressing" field was applied, this signal is necessarily due to a coherence transfer from Cs. Figure 2(e) represents the theoretical shape of the signal thus obtained at the output of the photomultiplier: The Rb signal starts from zero just when the rf field  $\tilde{H}_1 \cos \omega t$  is switched on, increases to a maximum value, and then decreases. It can be understood as resulting from a competition between the resonant dissipative spin-exchange coupling which makes coherence flow from Cs to Rb and the relaxation which causes the decay of both atomic orientations. Figures 3(a)-3(g) show the oscilloscope recordings of the experimental signal for different values of the

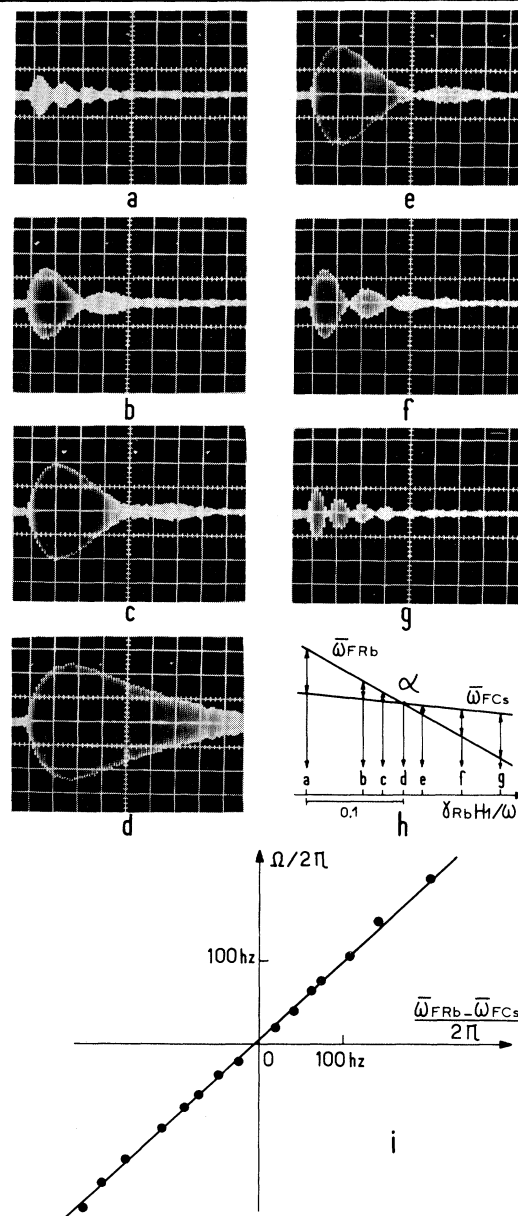


FIG. 3. (a)-(g): Oscilloscope recordings of the transfer signal for values of the field amplitude  $H_1$  around the first crossing point  $\alpha$  of Fig. 1. The  $H_1$  values corresponding to each picture are indicated in (h). The time scale of the recordings is 10 msec/division. In this experiment  $H_0 = 3.35$  mG ( $\omega_{\text{FRb}}/2\pi = 2350$  Hz);  $\omega/2\pi = 6400$  Hz. (d) corresponds to the resonant case. (h) Magnified drawing of the first crossing point  $\alpha$  in which the values of  $H_1$  corresponding to the pictures of (a)-(g) are indicated by arrows. (i) Plot of the beat note  $\Omega$  versus the mismatch frequency.  $\bar{\omega}_{\text{FRb}} - \bar{\omega}_{\text{FCs}}$  is calculated from the theoretical formula (2) giving the  $g$  factors of the "dressed" atoms.

field amplitude  $H_1$  around  $H_{1\alpha}$  [the  $H_1$  values corresponding to the different pictures are indicated by arrows on Fig. 3(h) which represents the first

crossing point  $\alpha$  of Fig. 1 magnified; Fig. 3(d) corresponds to the resonant case considered above]: One sees that the transverse angular momentum flows from Cs to Rb even if the “dressed” Larmor frequencies are slightly different, but the ac signal is now modulated at a frequency  $\Omega$ , which is to good approximation equal to the mismatch frequency as may be seen in Fig. 3(i) where we have plotted  $\Omega$  vs  $\bar{\omega}_{FRb} - \bar{\omega}_{FCs}$ . The modulation  $\Omega$  of the transient signal results from a beating between the coupled transverse angular momenta  $^F\bar{S}_+(Rb)$  and  $^F\bar{S}_+(Cs)$ .  $\Omega$  is equal to  $\bar{\omega}_{FRb} - \bar{\omega}_{FCs}$  because there are many more Cs atoms than Rb atoms in the sample so that the “feedback” of the Rb orientation on Cs is negligible. Otherwise, the correspondence between  $\Omega$  and  $\bar{\omega}_{FRb} - \bar{\omega}_{FCs}$  is more complicated and would also involve the various coupling constants and relaxation times. One can also notice in Figs. 3(a)-3(g) that the amplitude of the signal decreases as the mismatch frequency increases, the maximum transfer occurring at resonance [Fig. 3(d)].

All the experimental results described above may be qualitatively explained by a simple theoretical model, assuming only that the transverse angular momenta  $^F\bar{S}_+(Rb)$  and  $^F\bar{S}_+(Cs)$  on both alkali metals behave as two kinds of damped “oscillators” coupled together, whose eigenfrequencies  $\bar{\omega}_{FRb}$  and  $\bar{\omega}_{FCs}$  can be continuously varied by sweeping the rf field amplitude (see Fig. 1). The actual experimental situation is however somewhat more complicated for two different reasons: First, as we have already seen before, each alkali ground state has two hyperfine levels  $F$ ,  $F'$  of opposite Landé factors which are also coupled together via spin-exchange and wall-collision processes:  $B_1$  orients in fact both Cs hyperfine levels and  $B_2$  detects a linear superposition of the orientations in both Rb<sup>87</sup> levels, all these levels being coupled two by two with each other. One must thus take into account not only the coupling between upper  $F$  hyperfine levels, which is resonant for the crossing points  $\alpha$ ,  $\beta$ ,  $\gamma$ , ... of the solid-line curves of Fig. 1, but also the coupling between the upper  $F$  level of Rb<sup>87</sup> and the lower  $F'$  level of Cs<sup>133</sup>, for example, which must be resonant for the crossing points  $\alpha'$ ,  $\beta'$ ,  $\gamma'$ , ... of the dashed line  $\bar{\omega}_{F'Cs}$  curve with the solid-line  $\bar{\omega}_{FRb}$  curve. Thus the number of resonant values of  $H_1$  is doubled. Second, the interaction with the rf field does not only change the Landé factors of the levels but, as seen in Ref. 9, also modifies their eigenfunctions. Consequently, one

can show<sup>11</sup> that the Larmor precession of the different spins becomes anisotropic: Because of the preferential direction introduced by  $H_1$ , the motion of the spins is now elliptical in the plane perpendicular to  $H_0$ .  $^F\bar{S}_+(Rb)$  [ $^F\bar{S}_+(Cs)$ ] can be decomposed into two counter-rotating circular components evolving at frequencies  $\bar{\omega}_{FRb}$  and  $-\bar{\omega}_{FRb}$  ( $\bar{\omega}_{FCs}$  and  $-\bar{\omega}_{FCs}$ ). Moreover, the spin-exchange process becomes also anisotropic, the coupling being different for the spin components parallel and perpendicular to  $\vec{H}_1$ , so that all the clockwise and counterclockwise components of the spins are now coupled with each other in the different hyperfine levels. Therefore, when  $\bar{\omega}_{FRb} = -\bar{\omega}_{FCs}$ , resonances in the transfer must also occur between the  $\bar{\omega}_{FRb}$  circular component of the  $F$  level of Rb<sup>87</sup> and the  $-\bar{\omega}_{FCs}$  one of the  $F$  level of Cs; as  $\bar{\omega}_{F'Cs} = -\bar{\omega}_{FCs}$ , this process must also contribute to the  $\alpha'$ ,  $\beta'$ ,  $\gamma'$  resonances quoted above. All the resonances  $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\alpha'$ ,  $\beta'$ ,  $\gamma'$  have been experimentally observed. Their position as a function of  $H_1$  and the frequency of the corresponding transient signals are in good agreement with the theoretical values.

We will conclude with the following remark: For free atomic levels of different  $g$  factors, the transfer of coherence by collisions, which requires the equality of the atomic frequencies, exhibits a resonant variation when the static field  $H_0$  is swept around zero.<sup>7</sup> In the experiments described above, the coupling with a nonresonant rf field  $\vec{H}_1 \cos \omega t$  introduces for certain values of  $H_1$ , and whatever  $H_0$  is, equal Larmor frequencies in both atomic systems: The coherence transfer becomes independent of  $H_0$ , but exhibits sharp resonances when the amplitude of the rf field is varied, which is a quite unusual effect.

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## NUCLEAR MAGNETIC RESONANCE STUDIES OF $\text{Eu}^{151}$ AND $\text{Eu}^{153}$ IN EUROPIUM IRON GARNET SINGLE CRYSTALS

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The nuclear magnetic resonances of  $\text{Eu}^{151}$  and  $\text{Eu}^{153}$  have been studied in single crystals of europium iron garnet with externally applied fields of sufficient magnitude to align the magnetization along various crystallographic directions. Hyperfine-field and quadrupole-splitting parameters are obtained. Contributions to the electric field gradients and anisotropic hyperfine fields are discussed and compared with theory.

In recent years a number of experimental studies<sup>1-4</sup> using the Mössbauer effect as well as theoretical studies<sup>2-6</sup> have been carried out to determine the hyperfine fields and quadrupole interactions of the  $\text{Eu}^{+3}$  ions in europium iron garnet (EuIG). The Mössbauer studies, however, have been restricted to "zero fields" where the magnetization is along the easy,  $\langle 111 \rangle$ , direction. From Mössbauer data in this direction only, it turns out to be impossible to evaluate independently all three hyperfine-field parameters associated with the orthorhombic symmetry at the rare-earth site. We have studied<sup>7</sup> the NMR of  $\text{Eu}^{151}$  and  $\text{Eu}^{153}$  in a single crystal of EuIG with external fields sufficient to align the magnetization along the orthorhombic axes of the rare-

earth sites and have determined the hyperfine fields and quadrupole splittings.

The studies were made at 4.2°K using spin-echo equipment by plotting the spin-echo amplitudes as a function of frequency. The crystal, which was roughly spherical and about 1 cm in diameter, was mounted so that it could be rotated about the  $\langle 110 \rangle$  axis which was parallel to the rf field and perpendicular to the dc field  $H_0$ . Studies were made with a value of  $H_0$  of about 8 kG, sufficient to overcome the anisotropy field of EuIG which is about 5 kG at 4.2°K.<sup>8</sup> The  $\text{Eu}^{151}$  spectra for the case of  $H_0$  along the  $\langle 110 \rangle$  and  $\langle 100 \rangle$  directions are shown in Figs. 1(a) and 1(b). Corresponding  $\text{Eu}^{153}$  spectra are shown in Figs. 2(a) and 2(b). The spin-echo signals at each fre-

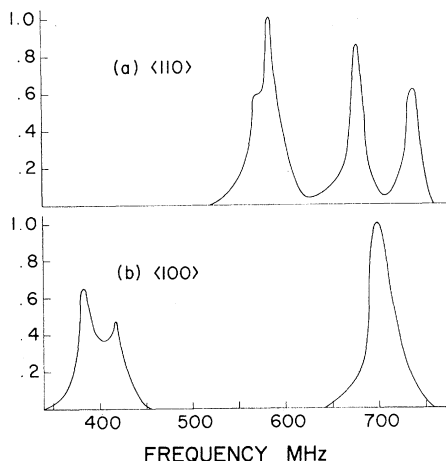


FIG. 1. (a) Corrected (see text) spin-echo spectrum of  $\text{Eu}^{151}$  at 4.2°K for  $H_0$  along the  $\langle 110 \rangle$  direction; (b) spectrum for  $H_0$  along the  $\langle 100 \rangle$  direction.

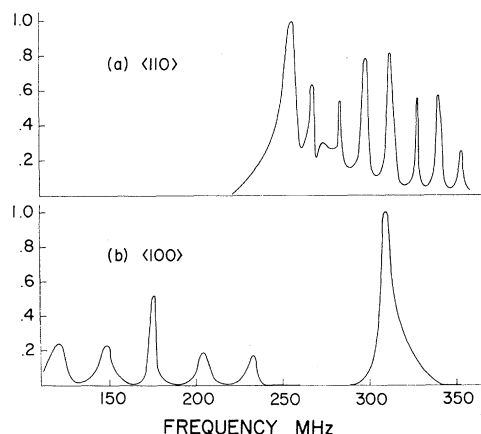


FIG. 2. (a) Spectrum of  $\text{Eu}^{153}$  at 4.2°K for  $H_0$  along the  $\langle 110 \rangle$  direction; (b) spectrum for  $H_0$  along the  $\langle 100 \rangle$  direction.

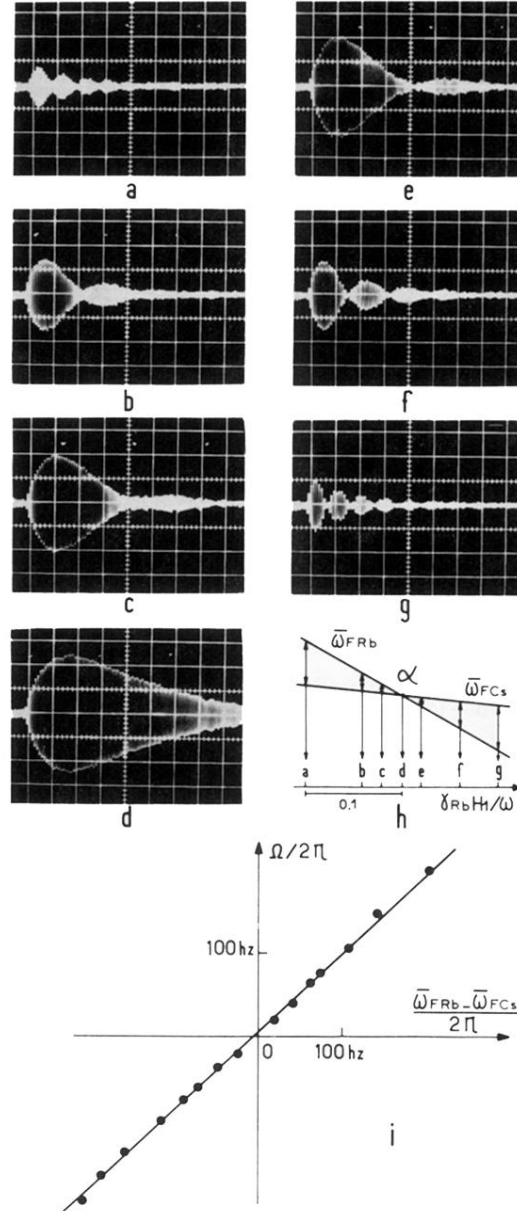


FIG. 3. (a)-(g): Oscilloscope recordings of the transfer signal for values of the field amplitude  $H_1$  around the first crossing point  $\alpha$  of Fig. 1. The  $H_1$  values corresponding to each picture are indicated in (h). The time scale of the recordings is 10 msec/division. In this experiment  $H_0 = 3.35$  mG ( $\omega_{FRb}/2\pi = 2350$  Hz);  $\omega/2\pi = 6400$  Hz. (d) corresponds to the resonant case. (h) Magnified drawing of the first crossing point  $\alpha$  in which the values of  $H_1$  corresponding to the pictures of (a)-(g) are indicated by arrows. (i) Plot of the beat note  $\Omega$  versus the mismatch frequency.  $\bar{\omega}_{FRb} - \bar{\omega}_{FCs}$  is calculated from the theoretical formula (2) giving the  $g$  factors of the "dressed" atoms.