high-field domain from the cathode is possible only between V_{crit} and V_2 and with currents above j_{crit}.

It has been observed that the applied voltage influences the domain width linearly, but does not influence the field strength in the domains. An increased optical excitation results in a reduction of the critical voltage. An increased additional IR quenching results in a decrease in the width of the high-field domain and an increase in critical voltage or, finally, at constant applied voltage, in a forced return to the homogeneous solution. Partial quenching of a layer parallel to the high-field domain results in a decrease in its width. A change in the sign of the net charge of the crystal with injecting contacts occurs as layer-like field inhomogeneities are initiated and as long as they remain adjacent to the cathode. All these experimental results are in good agreement with the proposed theory.

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"Zeeman" and "Dipolar" Spin Temperatures during a Strong rf Irradiation*

J. JEENER, R. DU BOIS, AND P. BROEKAERT†
Université Libre de Bruxelles, Bruxelles, Belgium
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Experiments have been performed on a single crystal of CaF_2 in a large constant magnetic field (7 kG), irradiated at the exact NMR frequency of F^{19} with transverse rf magnetic fields much larger than the local field. The results demonstrate the existence, in the steady state under very strong irradiation, of two independent quasi-invariants of the motion and of two corresponding temperatures which are the rotating-frame analogs of the usual laboratory-frame Zeeman and dipolar energies and temperatures in the absence of irradiation. Using pulse techniques, Zeeman and dipolar order have been transferred reversibly back and forth between the laboratory frame (without rf field) and the rotating frame (with the large rf field on). No loss of order or mixing between Zeeman and dipolar order has been observed for irradiation times up to 13 msec with a rotating component of the rf field of 25 G (the local field in CaF_2 amounts to a few gauss). The rotating-frame analog of the Strombotne-Hahn oscillations has also been observed.

IT has been shown by Redfield[1] that the properties of the spin system in a solid, in the presence of a large constant magnetic field $\mathbf{H}_0$ and of a transverse rf magnetic field $\mathbf{H}_1$, can be discussed in a frame of coordinates (X,Y,Z) rotating around $\mathbf{H}_0$ at the same angular velocity as $\mathbf{H}_1$, by means of an effective Hamiltonian H_{eff} which has the same structure as the Hamiltonian H in the laboratory frame (x, y, z with z and Z in the direction of $\mathbf{H}_0$) in the absence of rf irradiation.[2] Thus one anticipates very striking similarities between the behavior of spin systems in the presence or absence of rf irradiation, and many such similarities have been observed already. For instance, saturation by irradiation at the NMR frequency ($-\gamma\hbar\mathbf{H}_0$ or $-\gamma\hbar\mathbf{H}_{eff}$)[3] and free precession signals[4] have been observed in both cases.

When the Zeeman term of the Hamiltonian is not much larger than the spin-spin coupling term ($|\mathbf{H}_0|$ or $|\mathbf{H}_{eff}| \lesssim$ a few times the local field $|H_L|$), energy is exchanged rapidly between these two terms and one expects to reach a steady-state situation characterized by a single spin temperature. This conjecture has been very successful in the interpretation of adiabatic magnetization and demagnetization experiments in both reference frames.[5]

However, when the Zeeman terms of the Hamiltonian are much larger than the spin-spin coupling term, spontaneous energy flow between these terms becomes extremely slow, and a number of recent experiments have demonstrated that the dipolar energy in the laboratory frame is a quasi-invariant of the motion in

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† Predoctoral Fellow of the Institut pour l'Encouragement de la Recherche Scientifique dans l'Industrie et l'Agriculture

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[2] The fixed frame quantities $\mathbf{H}_0$ and H_D (spin-spin coupling Hamiltonian) are replaced in the rotating frame by $\mathbf{H}_{eff}$ (of the order of $\mathbf{H}_1$) and $H_D{}^0$ (that part of H_D which commutes with the fixed frame Zeeman Hamiltonian). Spin lattice relaxation will be ignored in the present discussion.

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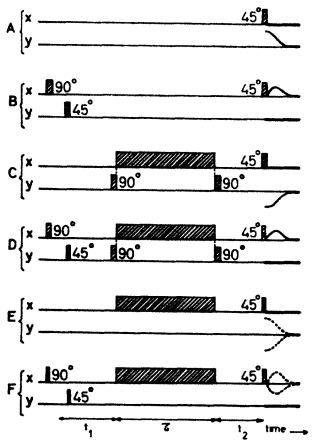

FIG. 1. X and Y components (in the rotating frame of coordinates) of the rf magnetic fields and nuclear magnetization in the various experiments. The angles shown correspond to the rf pulses. The free induction signals are indicated only where they were actually observed and measured: after the 45° information pulse. The time intervals t_1 and t_2 are much longer than T_2: a few hundred microseconds. We assume that the spin subsystems come to internal equilibrium during these intervals.

such cases and that a dipolar temperature can be used to describe this new type of order.[6] The experimental results reported in the present paper show that a similar situation exists in the case of strong rf irradiation when the rf magnetic field is much larger than the local fields.

The experiments have been done at room temperature on a single crystal[7] of CaF_2 (the only significant spin species in this crystal is F^{19}), containing 0.07% by weight of U^{+3}, at a field $\mathbf{H}_0$ of 7156 G, approximately oriented in a (100) direction, in times much shorter (a few milliseconds) than the spin lattice relaxation times in this field, which were, respectively, 1.14 sec for the Zeeman energy and 0.38 sec for the dipolar energy. Our rf pulse system generates linearly polarized rf magnetic fields at the exact NMR frequency (28.7 Mc/sec) of F^{19}, of adjustable amplitude, duration, and phase, homogeneous to within a few percent over the sample volume of 0.7 cm^3 (the component of the rf field rotating in the direction of the Larmor precession is of amplitude $\mathbf{H}_1$ and oriented along the X or the Y axis of the rotating frame). The free precession signals are processed by two mutually orthogonal phase-sensitive detectors, also oriented along the X and Y directions, which separate the components of the signals corresponding to Zeeman and dipolar order[8] [Figs. 1(A) and 1(B)]. For reasons of technical convenience we have replaced the standard method of transferring order from the Zeeman into the dipolar subsystem (preparation of states of low dipolar temperature by adiabatic demagnetization in the rotating frame[5,6]) by a phase-shifted double-pulse technique[9] which we have shown both theoretically and experimentally [Fig. 1(B)] to achieve the transfer with reasonable efficiency when the separation between pulses is of the order of T_2.

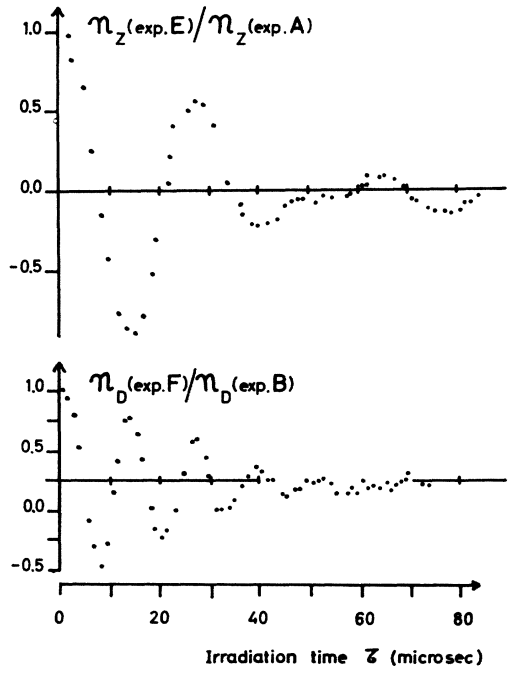

FIG. 2. Degrees of Zeeman and dipolar order (n_Z and n_D proportional to the amplitude of the corresponding components of the free precession signals) in experiments E and F, as a function of the irradiation time τ. The results shown all correspond to the same amplitude of rf field (rotating component of about 9 G). The quantities n_D(expt. E) and n_Z(expt. F) are very small.

Each experiment is performed on a spin system which is initially in thermal equilibrium with the lattice (Zeeman order).

Experiments A and B demonstrate the preparation and observation of states of (A) Zeeman and (B) dipolar order in the laboratory frame.

When the rf magnetic field is much larger than the local fields one expects a negligible coupling between the energies corresponding to the Zeeman term and to the secular part of the dipolar term of the effective Hamiltonian. In our experiments, the external magnetic fields appearing in H and in H_{eff} were, respectively, in the Z and X directions, and (assuming purely dipolar couplings) the secular part of the dipolar term of H_{eff} is equal to that of H, multiplied by $-1/2$ and rotated by 90° around the Y direction. The 90° pulse (around the Y direction) immediately followed by the sudden application of a large rf magnetic field in the X direction, which is used in experiments C and D, can thus be described as a simultaneous rotation by 90° around the Y direction of both the state of the spin system (90° pulse) and the Hamiltonian of interest. As a consequence of this, one expects that the degrees of order (or entropies) of the Zeeman and dipolar subsystems will be transferred without loss from the laboratory frame (with no rf field) to the rotating frame

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(with a large CW rf field on). The changes of magnitude of the Zeeman and secular dipolar terms of the Hamiltonian by factors of, respectively, $\mathbf{H}_{\text{eff}}/\mathbf{H}_0$ and $-1/2$ just cause irrelevant changes in the corresponding spin temperatures. Turning the CW rf field off after a time τ and immediately applying a 90° pulse around the Y axis result in similar rotations of the state and Hamiltonian of the spin system around the Y axis by, respectively, $+90°$ and $-90°$.

We have performed experiments C and D with a rotating component $\mathbf{H}_1$ of the rf field of about 25 G (the local fields in CaF_2 are of the order of a few gauss) and durations τ between 20 μsec and 13 msec (the free precession signal of CaF_2 dies out in about 50 μsec.) Our results are in complete agreement with the above predictions: The "90°–long pulse–90°" irradiation decreases the magnitude of the observed free precession signals by less than 13%; it changes the sign of the Zeeman signal and not that of the dipolar signal; the duration τ of the rf irradiation has no influence on the final state of the spin system; no mixing between Zeeman and dipolar energy in the rotating frame is observed.

Pulse sequence E was used to measure the magnitude of the rf magnetic fields from the frequency of the free precession in the effective field.

Experiment F is the rotating-frame analog of the Strombotne-Hahn experiment.[10] In the rotating frame, the spin system is first prepared in a state of order in zero effective field (dipolar order in the laboratory frame). A constant effective field is then turned on suddenly and, exactly as in the laboratory-frame experiment, the energy oscillates for a time of the order of T_2 back and forth between the Zeeman and the dipolar terms of the effective Hamiltonian, at about twice the Larmor frequency ω_1 in the effective field (the oscillations at ω_1 are absent in the case of strong irradiation at the exact NMR frequency ω_0). The sudden suppression of the effective field after a time τ destroys this Zeeman energy, but does not change the dipolar energy which then rearranges itself in the absence of rf field and is measured later on by means of the 45° information pulse. A straightforward calculation (see Ref. 8), using the hypothesis of two independent spin temperatures in the rotating frame, predicts that the laboratory-frame dipolar order recovered after a long and strong irradiation should be $\frac{1}{4}$ of the initial dipolar order, in good agreement with our experimental results.

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Electron-Spin-Resonance Spectrum of Mn^{2+} in β-Ga_2O_3*

V. J. FOLEN
U. S. Naval Research Laboratory, Washington, D. C.
(Received 8 April 1965)

The electron-spin-resonance spectrum of Mn^{2+} has been investigated in monoclinic β-Ga_2O_3 at 24 kMc/sec. It was found that the spectrum consists of a set of lines corresponding to a single type of environment. This spectrum (including the fine and $\Delta m=0$ hyperfine structure) is satisfactorily described by the spin Hamiltonian for Mn^{2+} in a rhombic crystal field with the following derived constants: $A_z=-87.7\pm0.2$ G; $A_y=-85.6\pm0.2$ G; $g_z=2.002$; $g_y=2.007$; $D=545.0$ G; $E=124.3$ G. The y crystal-field axis is along the monoclinic axis (b) and the z crystal-field axis makes an angle of 18° with the c crystal axis ($a>c$). Forbidden $\Delta m=\pm1, \pm2$ hyperfine transitions were also observed. From the separations between the $\Delta m=\pm1$ doublets, a value of $Q'=+0.9\pm0.2$ G was obtained. On the basis of the axial crystal fields inferred from the measured values of Q' in β-Ga_2O_3 and in other oxide materials, it appears that the calculated crystal-field contribution to the D parameter of Mn^{2+} does not explain the experimental results.

INTRODUCTION

THE origin of the ground-state splitting of the $3d^5-{}^6S_{5/2}$ ions (Mn^{2+} and Fe^{3+}) has been the object of a considerable number of theoretical and experimental investigations. Such investigations have indicated that these splittings may arise from the combined action of the spin-orbit interaction (W_{so}) and the crystal field,[1–5] or from the intra-ionic spin-spin interaction (W_{ss}) and the crystal field,[2,4–6] or from overlap and covalency between the S-state cation and its

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