[2]R. M. Sternheimer, Phys. Rev. 86, 316 (1952).
[3]R. M. Sternheimer, Phys. Rev. 95, 736 (1954).
[4]F. Herman and S. Skillman, Atomic Structure Calculations (Prentice-Hall, Inc., Englewood Cliffs, N. J., 1963).
[5]R. M. Sternheimer, Phys. Rev. 105, 158 (1957).

EXPERIMENTAL PROOF OF THE STRONG COUPLING BETWEEN THE ELECTRON SPIN-SPIN RESERVOIR AND A NUCLEAR SPIN SYSTEM IN DILUTE PARAMAGNETIC CRYSTALS

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Experiments are described in which the nuclear Zeeman temperature T_n and the temperature of the electron dipole-dipole interaction system $T_{\rm int}$ in a dilute paramagnetic crystal are measured simultaneously. The spin-lattice relaxation times of these two systems turn out to be equal, thus yielding a direct evidence of the strong coupling between these systems.

The concept of spin temperature in a rotating frame as introduced by Redfield[1] has led to a better understanding of the behavior of spin systems in solids subjected to strong rf fields. Redfield's theory, which was only valid in the limit of strong rf saturation, was extended by Provotorov[2] to rf fields of arbitrary intensity. He showed that in strong static magnetic fields two distinct temperatures should be assigned to the spin system: one temperature T_e for the Zeeman system and a temperature $T_{\rm int}$ to describe the dipole-dipole interaction system. He also derived equations for the time dependence of both temperatures under the influence of an rf field and of spin-lattice relaxation. Further calculations of the shape of the resonance absorption signal when Provotorov's concept of an interaction temperature $T_{\rm int}$ was taken into account were carried out by Rodak.[3] Atsarkin and Morshnev[4] performed line-shape experiments which confirmed the existence of a temperature $T_{\rm int}$ different from T_e. They were able to observe the time dependence of T_e and $T_{\rm int}$ by studying the shape of a paramagnetic resonance line which had been saturated by a slightly detuned microwave field.

An extension of these ideas to explain the results of nuclear dynamic polarization and nuclear relaxation experiments in dilute paramagnetic crystals provided further evidence for the existence of an interaction system of the paramagnetic ions. Actually measurements of the proton spin-lattice relaxation times in such crystals led to the idea that the nuclear Zeeman system could be strongly coupled to the interaction system of the paramagnetic ions.[5] Nuclear-dynamic-polarization experiments were carried out in order to confirm these views.[6] In performing these experiments however, only the nuclear resonance signal, which is inversely proportional to the nuclear Zeeman temperature T_n, was monitored. Thus only indirect information about the behavior of the interaction system was obtained.

In the experiments to be discussed below the nuclear Zeeman temperature T_n as well as the interaction temperature $T_{\rm int}$ were measured simultaneously in order to prove that T_n and $T_{\rm int}$ had the same time behavior, with a relaxation time equal to the nuclear spin-lattice relaxation time. This would be a direct proof of the strong coupling of the nuclear spins to the electron-spin interaction system.

The experiments were performed at 1.5 K in a single crystal of $La_2Mg_3(NO_3)_{12}0.24H_2O$ containing 2% Nd. The magnetic field H_0 of about 2.5 kG was directed perpendicular to the c axis. The paramagnetic resonance line of the Nd ions was observed at 9000 MHz, the proton resonance of the crystal waters occured at about 10 MHz.

The intensity of the proton resonance signal was used as a measure of the nuclear spin temperature T_n. The interaction temperature $T_{\rm int}$ was derived from the shape of the Nd resonance line. According to Provotorov[1] the rf absorption $\mathcal{P}(\Delta, t)$ of a spin system is given by

$$\mathcal{P}(\Delta, t) = P(\Delta)\left[\frac{\nu_e}{T_e(t)} + \frac{\Delta}{T_{\rm int}(t)}\right], \tag{1}$$

where $P(\Delta)$ is the transition probability due to the rf field and includes the usual line-shape function. ν_e is the Larmor frequency of the electron-spins. $\nu_e + \Delta$ is the frequency of the applied rf field. When both the Zeeman and the interaction system are in equilibrium with the lattice, Eq. (1) reduces to

$$\mathcal{P}(\Delta)=P(\Delta)\left[\frac{\nu_e}{T_L}+\frac{\Delta}{T_L}\right], \qquad (2)$$

where T_L is the lattice temperature. The antisymmetric term Δ/T_L can then be neglected compared with ν_e/T_L because $P(\Delta)$ is only different from zero for $\Delta \ll \nu_e$. The absorption line shape is thus symmetric about ν_e.

By applying a strong microwave field at a frequency $\nu_e+\Delta_1$, a considerable change of both T_e and $T_{\rm int}$ can be effected. After suddenly reducing this microwave field to a small nonsaturating value, the terms $\nu_e/T_e(t)$ and $\Delta/T_{\rm int}(t)$ will return to their equilibrium values ν_e/T_L and Δ/T_L with the time constants τ_{1e} and $\tau_{1e}{}^1$, respectively. So during the relaxation process the line shape is given by

$$\mathcal{P}(\Delta,t)=P(\Delta)\left[\left(\frac{\nu_e}{T_e(t)}-\frac{\nu_e}{T_L}\right)\exp(-t/\tau_{1e})+\frac{V_e}{T_L}+\left(\frac{\Delta}{T_{\rm int}(t)}-\frac{\Delta}{T_L}\right)\exp(-t/\tau_{1e}{}^1)+\frac{\Delta}{T_L}\right]. \qquad (3)$$

In the experiment first a strong microwave field was applied at a frequency $\nu_e+\Delta_1$, where Δ_1 was chosen such as to obtain a maximum cooling of the interaction system. Independent of whether a coupling between the interaction system and the nuclear Zeeman system exists, a theoretical description of the dynamic polarization process shows that for this value of Δ_1, also, a maximum enhancement of the nuclear resonance signal is obtained. In the present experiment this enhancement is about 50. Then the microwave field was suddenly reduced to the nonsaturating value, and the derivative of the paramagnetic resonance signal was recorded by slowly varying the external field H_0. The observed line shape turned out to be strongly asymmetric. This asymmetry gradually vanished only after several minutes (Fig. 1).

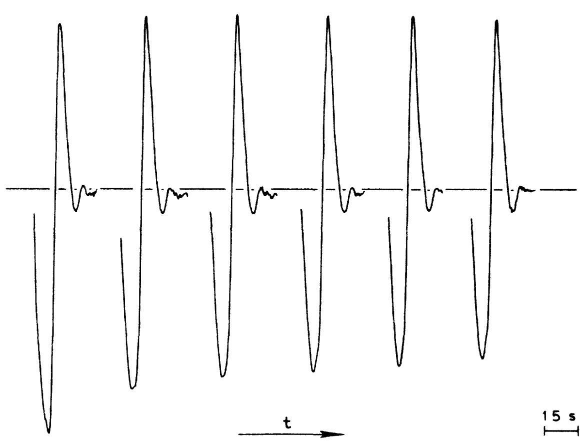

FIG. 1. Successive recordings of the Nd resonance line after strong cooling of the spin-spin interaction system. The asymmetry decays with a time constant of about 70 sec.

In order to measure the time constant involved, a second experiment was performed. After switching off the saturating field, the external field was adjusted at the center of the Nd resonance line ($\Delta=0$). As can be seen from Fig. 2 the derivative has a finite value at $\Delta=0$ directly after irradiation, which decays to zero (Fig. 3).[7] The time constant was measured to be 68 ± 2 sec at $T=1.5$ K. In this second experiment simultaneously a recording of the decay of the polarized proton signal was made. This signal also reduced to its equilibrium value with approximately

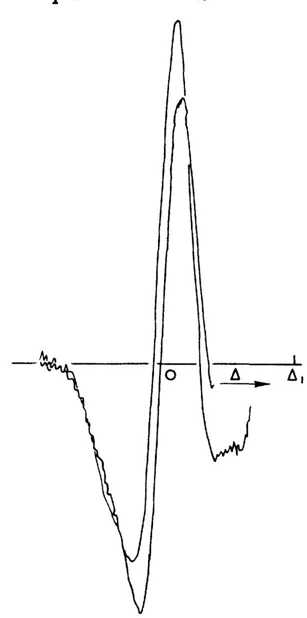

FIG. 2. Two recordings of the Nd resonance line; the asymmetric one is observed immediately after cooling of the interaction system by irradiation at $\nu_e+\Delta_1$. This asymmetric line decays to the more symmetric one.

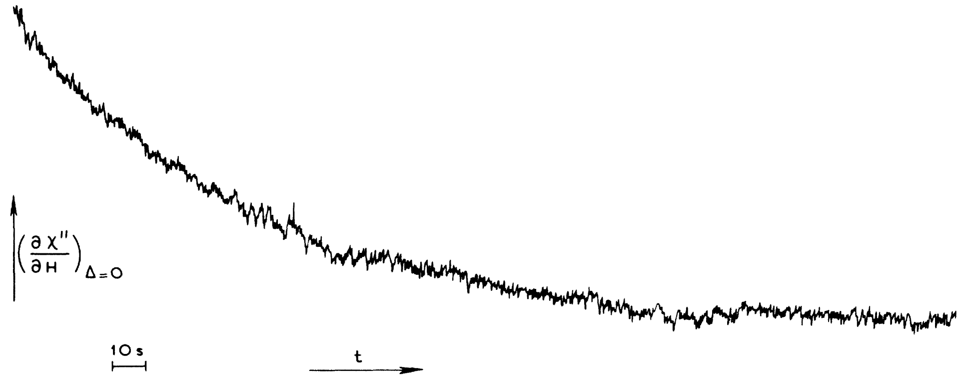

FIG. 3. Observation of the decay of the derivative of the resonance line at $\Delta = 0$. The measured time constant is 68 ± 5 sec.

the same time constant: 65 ± 3 sec.

This observation that the spin-lattice relaxation times of the nuclei and the interaction system are equal implies that these two subsystems are strongly coupled to each other.

If no such coupling existed, the relaxation time of the interaction system τ_{1e}[1] would be of the same order of magnitude, $\frac{1}{2}\tau_{1e}$,[1] as the Zeeman spin-lattice relaxation time τ_{1e}, which is about 0.5 sec at this temperature. The asymmetry of the Nd line should then have disappeared within a few seconds.

Further investigations in other crystals and under different experimental conditions are in progress and will be published in a forthcoming paper.

[1]A. G. Redfield, Phys. Rev. 98, 1787 (1955).

[2]B. N. Provotorov, Zh. Eksperim. i Teor. Fiz. 41, 1582 (1961) [translation: Soviet Phys.—JETP 14, 1126 (1962)].

[3]M. I. Rodak, Fiz. Tverd. Tela 6, 521 (1964) [translation: Soviet Phys.—Solid State 6, 409 (1964)].

[4]V. A. Atsarkin and S. K. Morshnev, Zh. Eksperim. i Teor. Fiz.—Pis'ma Redakt. [translation: JETP Letters 6, 288 (1967)].

[5]T. J. B. Swanenburg, thesis, Kamerlingh Onnes Laboratory, Leiden, The Netherlands, 1967 (unpublished); G. M. van den Heuvel, C. T. C. Heyning, T. J. B. Swanenburg, and N. J. Poulis, Phys. Letters 27A, 38 (1968).

[6]W. Th. Wenckebach, T. J. B. Swanenburg, H. Hoogstraate, and N. J. Poulis, Phys. Letters 26A, 203 (1968).

[7]The shift of the Nd resonance signal which is due to a change of the internal field caused by the polarization of the protons can be estimated to be an order of magnitude smaller than the effect observed here. Moreover, the symmetry of the line should not be affected by this internal field.

PROPAGATION OF STRONG-FIELD ELECTROMAGNETIC WAVES THROUGH PLASMAS NEAR THE ELECTRON CYCLOTRON FREQUENCY*

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The effect of electric field strength of the incident wave on the transmission of electromagnetic waves through an anisotropic plasma has been investigated using field strengths of 27.4 to 137 V/m. Near the electron cyclotron frequency increased transmission occurs for the strong field strengths.

Most investigations of electromagnetic wave interaction with plasmas are based on a "small-signal" approach where it is assumed that the electric field strength is too weak to affect appreciably the properties of the plasma. This approach has the advantage of simplicity, since the processes can be described by linear equations. Significant research has also been devoted to the other extreme case of very strong electromagnetic fields which can result in breakdown or in parametric excitation[1] of various wave frequencies.