CROSS SPIN RELAXATION IN THE HYPERFINE STRUCTURE OF ELECTRON PARAMAGNETIC RESONANCE OF Co^{2+} IN CORUNDUM

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Cross spin relaxation has been detected in the hyperfine structure of electron paramagnetic resonance of Co^{2+} ions in corundum. The cross-relaxation time T_{12} has been measured and found to be 0.27 sec and independent of the temperature.

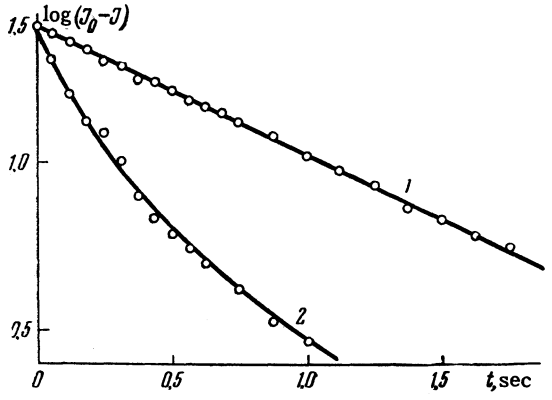
CROSS spin-spin interaction, i.e., interaction between spins with different resonance frequencies, plays an essential role in the dynamics of spin systems. In particular, it is of great significance in processes occurring in paramagnetic amplifiers.

The question of cross spin relaxation was recently analyzed in detail by Bloembergen et al.[1] who gave an explanation for the previously observed[2] effect of cross saturation of hyperfine structure components of electron paramagnetic resonance of copper ions in a single crystal of Tutton salt.

Cross-relaxation is prominent in systems of spins with not very different resonance frequencies. A group of closely spaced transitions occurs, for example, in the case of the Co^{2+} ion in corundum. For this ion the effective electron spin is $S' = 1/2$ and the nuclear spin is $I = 7/2$. The electron paramagnetic resonance lines for this ion have a well resolved hyperfine structure of eight components.[3,4]

We have investigated the cross spin relaxation of transitions corresponding to different orientations of the nuclear spin. The sample of corundum containing cobalt used by us had a low concentration of cobalt (10^{-2}%); the spin-lattice relaxation time T_1 at a temperature of 4.2°K was 1.2 sec. For the crystal orientation in the external magnetic field (H parallel to the trigonal crystal axis) used by us the width of an individual hyperfine structure component was 7.5 oe, the separation between the components was 30 oe.

The experiment was carried out in the following manner. The sample under investigation was placed into a rectangular resonator tuned simultaneously to two closely spaced frequencies ν_1 and $\nu_2 \sim 9200$ Mc/sec. The resonator had a nearly square cross section; oscillations of TE_{011} and TE_{101} type were excited in it. The sample was placed at a position of a maximum of the high frequency magnetic field in the resonator in such a way that the transitions between the levels would be induced by the high frequency field due to both types of oscillations. By means of a superheterodyne microwave spectroscope electron paramagnetic resonance lines were observed at the frequency ν_1 at a power level which excluded the possibility of saturation (10^{-10} w). Saturating pulses were applied at the frequency ν_2. The use of different frequencies for saturation and for observation enabled us to avoid interruption in the operation of the receiver due to the effect of the saturating pulse. The recovery of the intensity of the lines after the saturating pulse was turned off was recorded by a movie camera.



The diagram shows the dependence on the time of $\log(J_0 - J)$ for two different cases; J is the intensity of absorption which is proportional to the population difference between the spin levels n, while J_0 is the intensity of absorption in the case of thermal equilibrium.

In the case corresponding to curve 1 all eight hyperfine structure components were saturated to the same degree. Curve 2 corresponds to the case

when only one outermost component was saturated by a short pulse. In the first case the relaxation process is described by a single exponential

$$n_0 - n = A e^{-t/T_1}. \quad (1)$$

In the second case due to the cross spin-spin interaction the relaxation occurs more rapidly; the saturation of a single component immediately leads to partial saturation of other components, and only after a certain time the spin system regains equilibrium and relaxes as a whole according to the exponential law (1). The relaxation process in the second case can be easily calculated under the following assumptions: a) the cross-relaxation between each pair of neighboring components is described by the single parameter T_{12}, the cross-relaxation time, b) cross spin-spin interaction is taken into account only for neighboring components. In this approximation the relaxation of the populations of the eight pairs of spin sublevels differing in the values of the components of nuclear spin is described by the system of the following kinetic equations:

$$\begin{aligned}
\frac{dn_1}{dt} &= \frac{n_0 - n_1}{T_1} + \frac{n_2 - n_1}{T_{12}}, \\
\frac{dn_2}{dt} &= \frac{n_0 - n_2}{T_1} + \frac{n_3 - n_2}{T_{12}} + \frac{n_1 - n_2}{T_{12}}, \ldots \quad (2) \\
&\cdots\cdots\cdots\cdots\cdots \\
\frac{dn_8}{dt} &= \frac{n_0 - n_8}{T_1} + \frac{n_7 - n_8}{T_{12}}.
\end{aligned}$$

The solution of this system is a sum of eight different exponentials:

$$n_0 - n_i = \sum_{j=1}^{8} A_{ji} e^{-\lambda_j t}, \qquad \lambda_j = 1/T_1 + c_j/T_{12},$$

where the c_j are constants. The coefficients A_{ji} depend on the initial conditions.

Calculations show that the theoretical relaxation of a single hyperfine structure component agrees well with the experimental results if we take $T_{12} = 0.27$ sec. The results of such calculations are shown in the diagram by the curve, and the experimental values by points.

We note that the relaxation process at several different temperatures (1.8, 2.15, and 4.2° K) is characterized by different values of T_1 (3.0 sec at 1.8° and 1.2 sec at 4.2° K), while the same values are obtained for the parameter T_{12}.

In the case of saturation by a long pulse (of duration $\sim T_{12}$) the process is described by a similar system of kinetic equations, but the nature of the concrete relaxation process depends strongly on the amplitude and the duration of the saturating pulse.

[1] Bloembergen, Shapiro, Pershan, and Artman, Phys. Rev. **114**, 445 (1959).

[2] Giordmaine, Alsop, Nash, and Townes, Phys. Rev. **109**, 302 (1958).

[3] G. M. Zverev and A. M. Prokhorov, JETP **36**, 647 (1959) and **39**, 57 (1960), Soviet Phys. JETP **9**, 451 (1959) and **12**, 41 (1961).

[4] J. E. Geusic, Bull. Am. Phys. Soc. II, **4**, 261 (1959).

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