Contribution to the theory of stationary NMR signals under conditions of large dynamic frequency shifts

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The cause of the discrepancy between the theory of de Gennes *et al.* (Phys. Rev. **129**, 1105, 1963) and the experimental results of Portis *et al.* (J. Appl. Phys. **34**, 1052, 1963) and Tulin (Candidate's Dissertation, Moscow, 1970) is determined.

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One of the peculiarities of nuclear spin systems with dynamic frequency shift (DFS) is the dependence of the NMR frequency on the projection of the nuclear magnetization **m** on its equilibrium direction **z** (Ref. 1):

$$\tilde{\omega}_n = \omega_n - \omega_p m^z/m_0 , \qquad (1)$$

where ω_n is the NMR frequency without allowance for the DFS, ω_p is the DFS parameter which was of the order of 10–100 MHz in the experimental studies cited below,[2,3] and m_0 is the equilibrium value of **m**. This relation is the cause of the following singularity in the behavior of stationary NMR signals in transverse radiofrequency (RF) fields.

Let the frequency of the RF field be $\omega > \tilde{\omega}_n$ but $(\omega - \tilde{\omega}_n) < \omega_p$. Then, as shown in Fig. 1, besides the usual state corresponding to nonresonant small-amplitude force

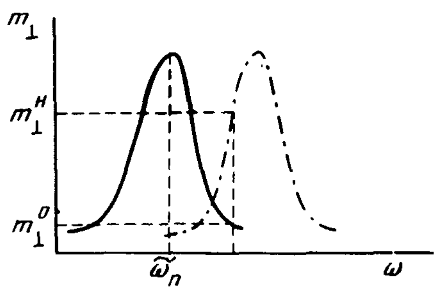

FIG. 1. Position of the NMR line for saturated (dash-dot) and unsaturated (solid) states of a nuclear spin system in a transverse RF field.

oscillations of $m_\perp^0$ (unsaturated state) there can exist a state with large oscillation amplitude $m_\perp^H$, corresponding to a shifted NMR line at a lower value of m^z (saturated state).

The existence of two stationary states of **m** in a transverse RF field was confirmed experimentally in Refs. 2 and 3. One of the methods of investigating such states involves observation of the antiferromagnetic resonance (AFMR) frequency, which also turns out to depend on m^z (Ref. 4), owing to the hyperfine interaction of the electron and nuclear spins. The shift of the AFMR frequency upon saturation of the nuclear

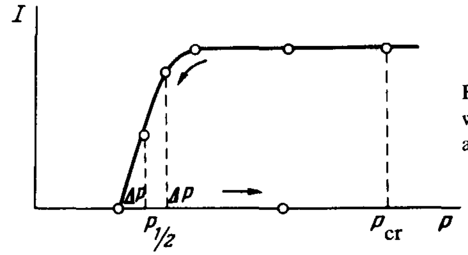

FIG. 2. The onset and vanishing of NMR saturation when the power of the saturating RF field is varied, in accord with AFMR data.[3]

spin system by the RF field is shown in Fig. 2, which is taken from Ref. 3.

The shifted AFMR line, corresponding to the saturated state **m** appears at a saturating RF field power $P = P_{cr}$. Once produced, it remains also at $P < P_{cr}$, and vanishes at $P < (P_{1/2} - \Delta P)$. In the interval $(P_{1/2} - \Delta P) \leqslant P \leqslant (P_{1/2} + \Delta P)$ there exists two AFMR lines, corresponding to saturated and unsaturated states of **m**. In Ref. 1 they analyzed the conditions for the existence of such two stationary states of **m** in a transverse RF field under the assumption that the NMR line shape is Gaussian. In this case the amplitude of the induced oscillations of **m** in a nonresonant RF field decrease exponentially with increasing detuning $(\omega - \widetilde{\omega}_n)$. The value obtained for the power P_{cr} in Ref. 1 was therefore exponentially large:

$$P_{cr} \sim \exp\{(\omega - \widetilde{\omega}_n)^2 / \Delta\Omega^2\}, \tag{2}$$

where $\Delta\Omega$ is the width of the NMR line. In the case of a Lorentz NMR line, the amplitude of the excited oscillations of **m** decreases with increasing detuning $(\omega - \widetilde{\omega}_n)$ in accordance with a power law, so that one should expect for P_{cr} values much smaller than given by (2). We advance below arguments that the NMR line shape for the considered case is better regarded as Lorentzian, and cite the results of the calculations of the values of P_{cr} and $P_{1/2}$.

As shown in Ref. 1, the DFS can be regarded as the result of correlations in the motion of the nuclear spins on account of the Suhl–Nakamura interaction $\mathscr{H}_{SN}$ (Ref. 5). It can be shown that these correlations suppress completely the inhomogeneities of the hyperfine fields at distances

$$\rho < r_0 , \tag{3}$$

where r_0 is the radius of the $\mathscr{H}_{SN}$ interaction, and is of the order of 10^{-5} cm for antiferromagnets of the easy-plane type.[4] Thus, the NMR frequencies for nuclear spins in a volume with dimensions $\rho \leqslant r_0$ (this volume contains up to 10^9 spins) can differ only on account of relaxation processes, i.e., the corresponding NMR line is homogeneously broadened.

The Gaussian NMR line shape corresponds to random scatter of the frequencies $\widetilde{\omega}_n$ (1) and is usually possessed by inhomogeneously broadened lines. Homogeneously broadened lines most frequently have a Lorentz shape, which describes the NMR line in the stationary regime, when the motion satisfies the Bloch equations.[6] In a coordinate frame that rotates together with a RF field these equations, take the form[6]:

$$\dot{m}^x = (\widetilde{\omega}_n - \omega) m^y - m^x / T_2 ,$$

$$\dot{m}^y = -(\widetilde{\omega}_n - \omega)m^x + \omega_1 m^z - m^y/T_2, \tag{4}$$

$$\dot{m}^z = -\omega_1 m^y + (m^z - m_0)/T_1,$$

where ω_1 is the amplitude of the RF field with allowance for the enhancement effect[4], in frequency units. In the stationary regime $m^\alpha = 0$ and the system (4) reduces to a single equation

$$\Delta z = -\omega_1^2 T_1 T_2 / [1 + \omega_1^2 T_1 T_2 + (\Delta\omega - \omega_p \Delta z)^2 T_2^2], \tag{5}$$

where $\Delta\omega = (\omega_n - \omega - \omega_p)$ and $\Delta z = (m^z - m_0)/m_0$.

Equation (5) is of the same form as the known equation for a nonlinear one-dimensional oscillator.[7] Its analysis leads to the following results. Three real roots (5) exist at values of $\Delta\omega$ in the interval

$$\Delta\omega_1 \leqslant \Delta\omega \leqslant \Delta\omega_2, \tag{6}$$

where the quantities $\Delta\omega_{1,2}$ at $\omega_1^2 T_1 T_2 \ll 1$ are given by:

$$\Delta\omega_1 = -\frac{1}{T_2}(\omega_p T_2 \omega_1^2 T_1 T_2), \tag{7}$$

$$\Delta\omega_2 = -\frac{3}{T_2}(\omega_p T_2 \omega_1^2 T_1 T_2/4)^{1/3}. \tag{8}$$

The product $\omega_p T_2$ is usually of the order 10^3–10^4, so that the inequality $\Delta\omega_{1,2} T_2 \gg 1$ can take place at $\omega_1^2 T_1 T_2 \ll 1$.

The smallest real root

$$\Delta z_1 = \Delta\omega/\omega_p \tag{9}$$

corresponds to a saturated state of the nuclear spin system, and the largest root

$$\Delta z_2 = -(\omega_1/\Delta\omega)^2 (T_1/T_2) \tag{10}$$

to an unsaturated state. It can be shown that the intermediate root corresponds to an unstable state of **m**. We point out that Δz_1 (9) is determined by the detuning $\Delta\omega$ and is independent, in first-order approximation, of the amplitude of the RF field ω_1.

The critical value of the power $P_{1/2}$ (Fig. 2) is determined by the value of $\Delta\omega_1$ (7):

$$P_{1/2} \sim \omega_1^2 = -\frac{\Delta\omega_1}{\omega_p}\frac{1}{T_1 T_2}, \tag{11}$$

and coincides with the value obtained in Ref. 1. For P_{cr} we get from (8)

$$P_{cr} \sim \omega_1^2 = \frac{4}{27}\frac{(-\Delta\omega_2 T_2)^3}{\omega_p T_2}\frac{1}{T_1 T_2} \tag{12}$$

This quantity increases in proportion to $\Delta\omega_2^3$, and not exponentially as found in Ref. 1 [see formula (2)].

The small experimental values of P_{cr} compared with the result of formula (2), were attributed in Refs. 2 and 3 to the influence of impurities. It was shown above that for Lorentz lines the value of P_{cr} (12) is quite small also in the case of ideal crystals. Nevertheless, the role of impurities in transitions between unsaturated and saturated states should be substantial, since such transitions are of first order and proceed via formation of germs of a new phase. The boundaries between the two phases are spatially inhomogeneous formations and a special study is needed for an analysis of their properties.

In conclusion, I am deeply grateful to V.A. Tulin for numerous discussions of this problem and for a number of valuable remarks, without which this work would hardly be possible.

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