Spin excitation kinetics in paramagnet by microwave field pulse

N Y Asadullina¹, T Y Asadullin¹

¹Kazan National Research Technical University named after A.N. Tupolev -KAI, Kazan, Russia
tyasadullin@kai.ru

Abstract. This work is devoted to the study of the kinetics of electron spins excitation in solid-state dilute paramagnets in the process of transient nutations. Transitional nutations are the response of a system, initially in thermal equilibrium, to a suddenly applied very intense resonance field during its transition to a new stationary state. The decay rate of the signal is determined by the interaction of active spins with each other $\Gamma_{ac}(\Delta, t) \sim n_{ac}(\Delta, t)$ and active spins with passive $\Gamma_{pas}(\Delta, t) \sim n_{pas}(\Delta, t)$. Therefore, the challenge is to find these concentrations. They are obtained by solving kinetic (rate) equations.

1. Introduction. The processes of creation, processing and transmission of quantum information include the excitation of a system of two-level particles-qubits with a sequence of pulses of a resonant coherent electromagnetic field of various duration and intensity. Such qubits can be nuclear or electron spins, impurity atoms or ions, quasiparticles such as magnons, excitons, polaritons, etc. Accordingly, the resonant frequencies $\omega$ of the exciting field extend from radio frequencies in the case of nuclear spins and microwaves (in the case of electron spins) to optical frequencies in the case of optical transitions in atoms and ions. As a result of quantum transitions between levels, the pulse field introduces coherent information into the system, which is read by the receiving device as a coherent response (signal). This response decays with one rate or another both during the action of the impulse and after it. Loss of information (relaxation) is a common characteristic of all the systems listed above and not named. Therefore, revealing the nature and properties of this relaxation (for each system) is one of the most important tasks of both theory and experiment.

Obviously, the signal value is proportional to the number (concentration) of particles in the system. If the other characteristics of the signal, in particular, the decay rate $\Gamma$, did not depend on the concentration, it could be ignored, since the signal is measured in relative units. However, experience shows that the decay rate depends on both the amplitude $B_1$ of the pulse field $2B_1 \cos(\omega t)|\mathbf{x}$ and the concentration $n$: $\Gamma = \Gamma(B_1, n)$. The total concentration $n$ consists of the concentration $n_{ac}(t)$ of the "active" particles excited by the pulse and the "passive" $n_{pas}(t)$: $n = n_{ac}(t) + n_{pas}(t)$. Naturally, the magnitude of the signal is proportional to $n_{ac}(t)$; however, both concentration components contribute to the damping of the response. In turn, the concentration of active particles itself depends on the intensity of the pulse field: $n_{ac}(B_1, t)$. It is reasonable to assume that the contributions to the decay rate from active $\Gamma_{ac} \sim n_{ac}(t)$ and passive $\Gamma_{pas} \sim n_{pas}(t)$ components can be caused by different mechanisms.
This work is devoted to the study of the kinetics of the excitation of electron spins in solid diluted paramagnets in the process of transient nutations. Transitional nutations are the response of a system, initially in thermal equilibrium, to a suddenly applied very intense resonance field during its transition to a new stationary state. This phenomenon was originally discovered in the system of nuclear spins [1], then it was observed in the system of electron spins [2], on optical transitions, in atomic and molecular systems [3]. More specifically, our goal is to describe the experiments of Boscaino and co-workers [4, 5] on the attenuation of nutations in quartz and amorphous silicon.

The materials under consideration are characterized by inhomogeneous broadening, that is, the spread of resonance frequencies of spins (spin packets) ω0j relative to the central frequency ω0, which coincides with the frequency of the pulse field: ω0 = ω. Usually this spread is approximated by a Gaussian distribution

\[ f(Δ) = \left(\frac{2π}{σ}\right)^{-1/2} e^{-Δ^2/2σ^2}, \]

with a standard deviation σ. In addition, the resonance transition for each spin packet is uniformly broadened. In our case of saturation by a strong field, this broadening is described by the Lorentz distribution

\[ L(Δ) = \frac{1}{π} \frac{Γ_0}{Δ_0^2 + Γ_0^2}, \]

where \( s_0 = χ^2 T_2^2 \) is the saturation parameter. Here \( χ = γB_1 \) is the Rabi frequency, \( γ \) is the gyromagnetic ratio, \( Γ_0 = 1/T_2 \) is the relaxation rate at a weak excitation field.

As mentioned above, we have signal decay rates due to the interaction of active spins with each other \( Γ_{ac}(Δ, t) \sim n_{ac}(Δ, t) \) and active spins with passive \( Γ_{pas}(Δ, t) \sim n_{pas}(Δ, t) \). Therefore, the challenge is to find these concentrations. They are obtained by solving kinetic (rate) equations. The system of spins in thermal equilibrium is characterized by the populations \( n_{10} \) of the lower and \( n_{20} \) upper levels: \( n = n_{10} + n_{20} \). The equilibrium spectral concentrations will be \( n_{10}(Δ) = n_{10}(Δ) \) and \( n_{20}(Δ) = n_{20}(Δ) \), respectively. During the pulse, the dependence of the spectral concentrations on time \( n_{10}(Δ, t) \) and \( n_{20}(Δ, t) \) is described by the rate equations

\[ \frac{dn_{10}(Δ, t)}{dt} = -W_{12} n_{10}(Δ, t) - W_{21} n_{20}(Δ, t), \]

\[ \frac{dn_{20}(Δ, t)}{dt} = W_{12} n_{10}(Δ, t) + W_{21} n_{20}(Δ, t). \]

Here \( W_{12} \) and \( W_{21} \) are the probabilities of relaxation transitions and \( W_{12} = W_{21} = W \) are the probabilities of induced transitions. The probability of spontaneous transitions in the microwave frequency region is small and is not taken into account here.

We consider in a simplified way the real process of nutation excitation as consisting of several separate processes. Similar process [5] is the oscillations gradually decreasing in amplitude. It lasts several tens of microseconds. However, the first oscillation, the largest in amplitude, appears already in the first microseconds after the application of the pulse. Therefore, in the case of an intense field in the initial section, we neglect small relaxation terms. For example, in [5], the longitudinal relaxation time \( T_1 = 1/(W_{12} + W_{21}) \) is of the order of \( T_1 ≈ 1 \) second. Therefore:

\[ \frac{dn_{10}(Δ, t)}{dt} = -W (n_{10}(Δ, t) - n_{20}(Δ, t)); \]
\frac{d}{dt} n_z(\Delta,t) = -W\left(n_z(\Delta,t) - n_t(\Delta,t)\right) \tag{4b}

Here [6] \( W = \frac{\chi^2}{2a} \sin(at) / 2a, \ a = \sqrt{(\omega - \omega'_0)^2 + \chi^2}, \ \omega'_0 \) is the uniformly broadened frequency of the spin packet \( \omega_0 \). This transition probability should be averaged over the uniform broadening

\[ W(t) = \frac{\chi^2}{2} \int \frac{\sin(at)}{a} L(\omega_0') d\omega_0' \tag{5} \]

In the case of a short time of interest to us, the width of the \( \sin(at)/a \) function is much larger than the width of the Lorentzian function. Therefore, the function \( \sin(at)/a \) can be removed from the integral sign at the value \( a = \chi \).

Therefore, we finally have \( W(t) = (\chi/2) \sin(\chi t) \).

The solution to equations (4) has the form

\[ n_z(\Delta,t) = \frac{1}{2} [n_0 - \Delta n_0 \exp(\cos \chi t - 1)] f(\Delta); \quad \Delta n_0 = n_{10} - n_{20}; \tag{6} \]

\[ n_{ac}(\Delta,t) = [n_z(\Delta,t) - n_{20}(\Delta)]. \tag{7} \]

By integrating \( n_{ac}(\Delta,t) \) over the inhomogeneous spectrum, we find the total concentration \( n_{ac}(t) \) of active spins due to induced transitions.

**Conclusion.** The obtained solutions (6) and (7) describe well the initial stage of nutation oscillations [4, 5, 7] and in all respects are appropriate for describing echo phenomena [8, 9], which are known to be excited by very short pulses. A complete description of the entire nutation process is possible only by (numerical) solution of equations (3a), (3b) and relation (5).

**References**

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