About Excitation of a Quadrupole Spin Echo

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For the first time the excitation of a quadrupole spin echo by a sequence of radio frequency pulses with the filling frequencies ω_Q and $\omega_Q \pm \Delta\omega_Q$ is theoretically and experimentally considered, where ω_Q is the resonance frequency of the raised transition and $\Delta\omega_Q$ the offset within the half-width of the NQR line. It is shown that in this case the amplitude of the observable signals does not depend on the offset size, and the echoes appear at times which depend on the intervals between pulses, on the ratio $\Delta\omega_Q/\omega_Q$, and on the offset sign.

The experimental observation [1] and the theory [2] of a quadrupole spin echo assume a periodic influence of radiofrequency (r. f.) pulses on a sample containing quadrupole nuclei, where the filling frequency is equal to the resonance frequency ω_Q of the raised transition and where the echo registration occurs also at this frequency.

In [3] a nuclear spin-system was experimentally studied under the influence of r. f. pulses with the filling frequency $\omega_Q \pm \Delta\omega_Q$, where $\Delta\omega_Q$ is the offset within the limits of the NQR line half-width, and the registration of the response is performed for this frequency too.

In the present work the r. f. pulse sequences with filling frequencies equal to ω_Q and $\omega_Q \pm \Delta\omega_Q$ are considered. The registration of the echo signals is conducted on the resonance frequency.

Let us consider two (from many possible) variants of three-pulse excitation of the stimulated echo.

In the first variant the first radio frequency pulse is applied with the filling frequency ω_Q and at the times τ_1 and τ_2 the second and third radio frequency pulses follow with the filling frequency $\omega_Q + \Delta\omega_Q$.

In the second variant the first radio frequency pulse is applied with the filling frequency ω_Q, at the time τ_1 the second pulse follows with the filling $\omega_Q \pm \Delta\omega_Q$, and at the time τ_2 the third radio frequency pulse follows with the filling frequency ω_Q - $\Delta\omega_Q$. The size of $\Delta\omega_Q$ is always the same. In both variants the echo signal registration is carried out on the resonance frequency ω_Q.

In case of the first variant the echo signals are observed with the amplitudes

$$E^{(1)}_{m,m-1} = 2(I'_x)_{m,m-1} c_1(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_1)], \tag{1}$$

where $t_1 = (2 + \Delta\omega_{m,m-1}/\omega_{m,m-1})\tau_1$;

$$E^{(2)}_{m,m-1} = 2(I'_x)_{m,m-1} c_2(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_2)], \tag{2}$$

where $t_2 = \left(2 + \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_1 + \left(1 + \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_2$;

$$E^{(3)}_{m,m-1} = 2(I'_x)_{m,m-1} c_3(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_3)], \tag{3}$$

where $t_3 = \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\tau_1 + 2\left(1 + \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_2$;

$$E^{(4)}_{m,m-1} = 2(I'_x)_{m,m-1} c_4(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_4)], \tag{4}$$

where $t_4 = \left(1 + \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_1 + 2\left(1 + \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_2$;

$$E^{(5)}_{m,m-1} = 2(I'_x)_{m,m-1} c_5(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_5)], \quad (5)$$

where $t_5 = \left(2 + \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_1 + 2\left(1 + \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_2$.

In case of the second variant the echo signals are observed with the amplitudes

$$E^{(1)}_{m,m-1} = 2(I'_x)_{m,m-1} c_1(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_1)], \quad (6)$$

where $t_1 = \left(2 - \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_1 - 2\frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\tau_2$;

$$E^{(2)}_{m,m-1} = 2(I'_x)_{m,m-1} c_2(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_2)], \quad (7)$$

where $t_2 = \left(2 - \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_1 + \left(1 - \frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\right)\tau_2$;

$$E^{(3)}_{m,m-1} = 2(I'_x)_{m,m-1} c_3(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_3)], \quad (8)$$

where $t_3 = -\frac{\Delta\omega_{m,m-1}}{\omega_{m,m-1}}\tau_1 + 2\tau_2$;

$$E^{(4)}_{m,m-1} = 2(I'_x)_{m,m-1} c_4(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_4)], \quad (9)$$

where $t_4 = (1 - \Delta\omega_{m,m-1}/\omega_{m,m-1})\tau_1 + 2\tau_2$;

$$E^{(5)}_{m,m-1} = 2(I'_x)_{m,m-1} c_5(x_i)\omega_{m,m-1} \cdot \sin[\omega_{m,m-1}(t - t_5)], \quad (10)$$

where $t_5 = (2 - \Delta\omega_{m,m-1}/\omega_{m,m-1})\tau_1 + 2\tau_2$.

Here $(I'_x)_{m,m-1}$ is the operator matrix element of the operator I_x in the quadrupole hamiltonian representation $\mathcal{H}_Q$, $c_i(x_i)$ are trigonometrical functions of the flip angles of the radiofrequency pulses, $\omega_{m,m-1}$ is the resonance frequency ω_Q of the raised transition, $\Delta\omega_{m,m-1}$ the offset from the resonance frequency within the limits of the NQR line half-width, τ_1 and τ_2 are the time intervals between the exited pulses, and m is the magnetic quantum number.

Such three-pulse excitations have the property that the signal amplitudes practically do not depend on the offset size. The signal sites depend on the time intervals τ_1 and τ_2 (as in [1, 2]) between the exited pulses, and in addition on the ratio $\Delta\omega_{m,m-1}/\omega_{m,m-1}$ and on the offset sign.

At $\Delta\omega_{m,m-1} \to 0$ we receive the expressions (1) - (10) for the amplitudes and the echo signals sites as presented in [2].

The experimental observation has been carried out with a multifrequency pulse NQR spectrometer on ^{63}Cu in $Y_1Ba_2Cu_3O_{7-d}$ ($d > 0$), the resonance frequency being 31.12 MHz ($T = 297$ K). The width of this NQR line is ca. 200 kHz. It is necessary for the offset size to be established within less than 100 kHz. At large values of τ_1 and τ_2, shifts are observed in the echo signal sites (with respect to Hahn type signals).

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