Multispin magnetic resonance with the participation of polarized β-active nuclei

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It is found that impurity beta-active nuclei ^{8}Li in LiF undergo resonant depolarization not only when the specimen is irradiated by a radio-frequency field, rotating with the Larmor frequency of ^{8}Li, but also at frequencies that are linear combinations of the Larmor frequencies of ^{8}Li, ^{7}Li, and ^{19}F with integer coefficients. The frequencies, forbiddenness parameters, and second moments of the resonances are measured.

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The application of strong radio-frequency fields in magnetic resonance on impurity polarized β-active nuclei (β nuclei) allows observing a number of interesting phenomena in solids. For example, multiquantum transitions between quadrupole split levels of β nuclei were studied in Ref. 1; a very narrow NMR line with a width of about 30 Hz was obtained in Ref. 2; the line shape far from the center of the resonance, where its magnitude was 10^{-5} of the value at the center, was measured in Ref. 3. In this paper, we report the observation of resonance depolarization of ^{8}Li β nuclei, which participate in simultaneous reorientation of a group of spins in a polycrystalline specimen of LiF. The resonant frequencies ν_{0i} satisfy the conditions

$$\nu_{0i} = n\nu_8 + m\nu_{19} + k\nu_7\,,\ n \neq 0, \qquad (1)$$

where n, m, and k are integers; ν_8, ν_{19}, and ν_7 are the Larmor frequencies of ^{8}Li, ^{19}F, and ^{7}Li nuclei. It is natural to refer to these resonances as *multispin resonances*, since they correspond to transitions with simultaneous change in the magnetic quantum numbers of several spins.

The method for observing the resonances was based on the fact that the angular distribution of β emission of a polarized nucleus is related to the polarization $P(t)$ by the relation $W(\theta) \sim 1 + \text{const}\,P(t)\cos\theta$ where θ is the angle between the direction of polarization of the nucleus and the escape direction of the β particle. The $(0-\pi)$ angular asymmetry

$$\epsilon = [N(0) - N(\pi)]/[N(0) + N(\pi)]$$

of β emission of ^{8}Li nuclei ($T_{1/2} = 0.84$ s, spin $I = 2$), formed in the polarized state in the reaction $^{7}\text{Li}(n,\gamma)^{8}\text{Li}$ induced by thermal polarized neutrons [$N(\theta)$ is the number of

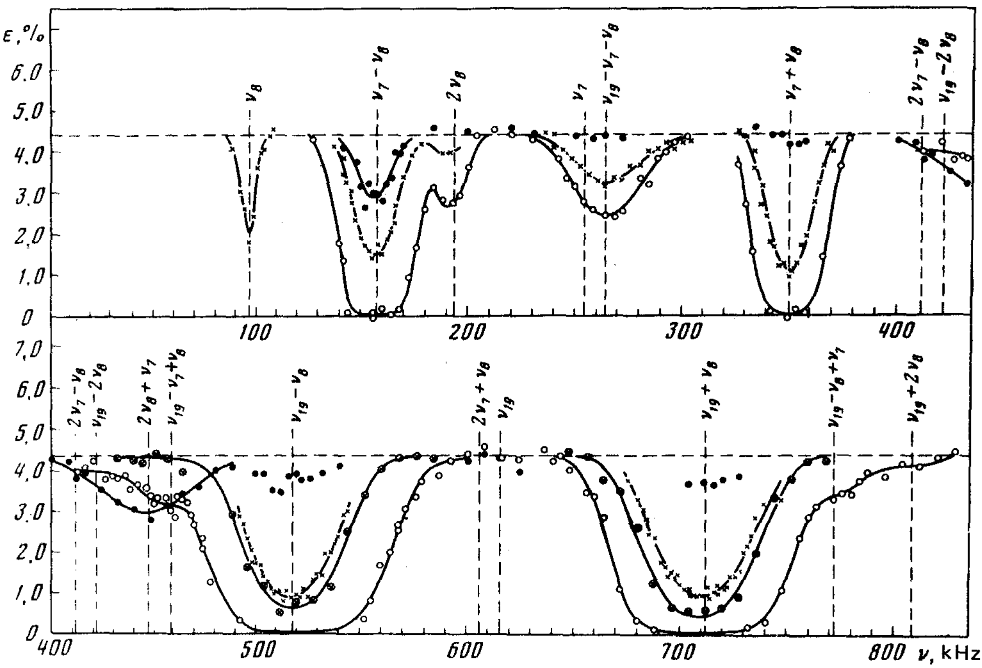

FIG. 1. The angular asymmetry of β emission of ^{8}Li nuclei as a function of frequency ν of the rf field. The frequencies ν_8, ν_7, and ν_{19} are the Larmor frequencies of the ^{8}Li, ^{7}Li, and ^{19}F nuclei. The points ○ were measured with an rf field rotating in the direction of precession of the ^{8}Li, ^{7}Li, and ^{19}F nuclei (their g factors are positive); the amplitude of the field H_1 was 6–8 G in the frequency range 130–400 kHz and 3–4 G in the range 400–800 kHz. The points ● were measured with the same rf field amplitudes H_1, but with field rotating in the opposite direction. The points × and ⊗ were measured with the direction of rotation of H_1 the same as that of precession of the nuclei, but with smaller amplitudes H_1: $H_1 = 0.02$ G for the ν_8 resonance; $H_1 = 1.7$ G for the $\nu_7 \pm \nu_8$ resonances; $H_1 = 5$ G for the $\nu_{19} - \nu_7 - \nu_8$ resonance; $H_1 = 0.6$ G and $H_1 = 0.9$ G for the $\nu_{19} \pm \nu_8$ resonances. The resonance near 450 kHz with the opposite direction of rotation of H_1 is related to resonance depolarization of the neutron beam by the rf field (the g factor of the neutron is negative). The absolute statistical accuracy of a single measurement is $\pm 0.14\%$.

counts on the counter, recording the decay electrons] was measured. The LiF specimen was held at room temperature, and the content of ^{6}Li isotopes was less than 0.3%.

We studied the dependence of ϵ on the frequency ν, amplitude H_1, and direction of rotation of the rf field. The field $H_0 \simeq 150$ G and the amplitude H_1 varied in the range 0.6-10 G. The resonances, shown in Fig. 1, were observed in the frequency range from 130 to 830 kHz. In analyzing the results, we assumed that the rate of depolarization of a β nucleus under the action of an rf field R is described by the equation

$$R = \pi \gamma_8^2 H_1^2 \sum_i A_i f_i \,(\omega - \omega_{0i}), \qquad (2)$$

where $\omega_{0i} = 2\pi\nu_{0i}$ is the frequency of the ith resonance, A_i is the forbiddenness parameter, $f_i(\omega)$ is the normalized function of the resonance shape, $\gamma_8 = 3.96 \times 10^3\ \mathrm{G^{-1}s^{-1}}$ is the gyromagnetic ratio of ^{8}Li nucleus. It was assumed that $f_i(\omega)$ is Gaussian with second moment $(2\pi)^2 M_{2i}$. The results of the analysis are presented in Table I.

It is well known that an isolated nucleus has only a single resonance frequency, the Larmor frequency. A multispin resonance arises because the combination frequencies (1) occur in the spectrum of collective spin motions due to the superposition of Zeeman and nonsecular dipole-dipole interactions and selection rules of the type $\Delta I_z = 1$ are removed. Thus, for example, only a single nucleus participates in the resonance at the frequency $2\nu_8$, since the concentration of β nuclei in the specimen is very low ($< 10^{15}$). However, this resonance, essentially a multispin resonance, cannot occur without the participation of the spins in the crystal.

The theory of multispin resonance can be constructed based on ideas similar to those that are used in the modern theory of multipulse NMR methods[4,5] and in nonlinear mechanics.[6,7] In constructing the effective Hamiltonian $\mathcal{H}_e$, it is convenient to go over to the interaction representation, choosing as the Hamiltonian $\mathcal{H}_0$ the interaction of all spins in the external field ($H \lessapprox H_{\mathrm{loc}}$, H_{loc} is the local field), and then to eliminate the rapidly oscillating terms, using unitary transformations[4,5] and an Arnold iteration procedure.[6,7] The effect of slowly oscillating terms from $\mathcal{H}_3$ on the depolarization of the nuclei is taken into account using the standard perturbation theory. In order to estimate the widths of the resonances, the standard approximation is used. In this approximation the correlation of local fields on different nuclei is ignored. The theory shows that reson-

TABLE I. Results of Analysis of the NMR Spectra.

	$\nu_7 - \nu_8$	$\nu_7 + \nu_8$	$\nu_{19} - \nu_8$	$\nu_{19} + \nu_8$	1) $-(\nu_7 - \nu_8)$	$\nu_{19} - (\nu_7 + \nu_8)$	$\nu_{19} + (\nu_7 - \nu_8)$	$\nu_{19} - (\nu_7 - \nu_8)$	$2\nu_8$
$A_i \cdot 10^3$	0,67 ±0,03	0.85 ±0.03	13,8 ±0,4	14,3 ±0,4	0,017 ±0,002	0,030 ±0,005	0,07 ±0,02	0,03 ±0,02	0,009 ±0,001
M_{2i}, kHz^2	57 ± 4	52 ±3	203 ±9	244 ±10	49 ±7	157 ±23	210 ±110	102 ±45	46 ±8

1)The minus sign indicates that the resonance is caused by an rf field rotating in a direction opposite to that of precession of the ^{7}Li and ^{8}Li nuclei.

ance must occur when the condition

$$l\nu_{0i} = n\nu_8 + m\nu_{19} + k\nu_7, 1 \leqslant |n| \leqslant 2I \qquad (3)$$

is satisfied. Small shifts in the Zeeman frequencies, with a relative order of magnitude $(H_{loc}/H_0)^2$, are omitted in this equation. The general tendency is such that as the order of the resonance increases, its forbiddenness parameter decreases.

The series of results calculated using the method presented above agrees well with experiment: Eq. (1) is reproduced; the correct values are obtained for estimates of the forbiddenness parameters and the second moment of the resonances; the ratio of the forbiddenness parameters calculated in the lowest order for two spin resonances coincide almost exactly with experiment: $A(\nu=\nu_7+\nu_8){:}A(\nu=\nu_7-\nu_8){:}A(\nu=\nu_8-\nu_7)$ $=(1.28{:}1{:}0.018)_{\text{theory}} = [(1.27\pm0.04){:}1{:}(0.017\pm0.02)]_{\text{exp}}$; $A(\nu=\nu_{19}+\nu_8){:}A(\nu=\nu_{19}-\nu_8){:}A(\nu=\nu_8-\nu_{19})=[1.05:1:(6\cdot10^{-4})]_{\text{theory}} = [(1.04\pm0.06):1:(<0.06)]_{\text{exp}}$.

The theory explains the reason for the narrowness of the resonance at the frequency $2\nu_8$. The line-shape function of this resonance is determined by the correlation function, constructed from the z component of ^{19}F spins, rather than from the x and y components, as for the resonance $\nu=\nu_{19}\pm\nu_8$, for example. The flip-flop process in LiF occurs much more slowly than phase relaxation, so that the resonance $\nu=2\nu_8$ is much narrower than the resonance $\nu=\nu_{19}\pm\nu_8$.

Multispin resonant transitions can also be observed using standard methods, without the use of β nuclei. In particular, the method of dynamic polarization of nuclei is based on the use of the simplest multispin transitions: two-spin transitions.[8] Multispin processes in systems of nuclear spins were recently observed with the use of pulsed NMR methods.[9]

The phenomenon, discovered and studied in this work, allows studying with the help of nuclear detection methods NMR spectra with the participation of stable isotopes in the specimen; for example, it allows studying the quadrupole interactions of these isotopes with radiation defects, which arise with the formation of β nuclei in crystals in (n,γ) reactions. The method could also be useful for studying local characteristics of spin-kinetic processes.

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1. D. Dubbers, K. Dörr, H. Ackermann, F. Fujara, H. Grupp, M. Grupp, F. Heitjans, A. Körblein, and H. J. Stöckmann, Z. Physik A **282**, 243 (1977).
2. A. Winnacker, D. Dubbers, F. Fujara, K. Dorr, H. Ackermann, H. Grupp, P. Heitjans, A. Körblein, and H. J. Stöckmann, Phys. Lett. A **67**, 423 (1978).
3. M. I. Bulgakov, A. D. Gul'ko, Yu. A. Oratovskiĭ, and S. S. Trostin, Zh. Eksp. Teor. Fiz. **61**, 667 (1971) [Sov. Phys. JETP **34**, 356 (1972)].
4. B. N. Provotorov and É. B. Fel'dman, Zh. Eksp. Teor. Fiz. **79**, 2206 (1980) [Sov. Phys. JETP **52**, 1116 (1980)].
5. L. L. Bushivili, E. B. Volzhan, and M. G. Menabde, TMF **46**, 251 (1981).
6. N. N. Bogolyubov and Yu. A. Mitropol'skiĭ, Asymptoticheskie metody v teorii nelineĭnykh

kolebaniĭ [Asymptotic Methods in the Theory of Nonlinear Oscillations], Nauka, Moscow (1974), Section 28.
7. V. A. Arnol'd, Matematicheskie metody klassicheskoĭ mekhaniki [Mathematical Methods of Classical Mechanics], Nauka, Moscow (1974), Chapter 10.
8. K. Jeffis, Dynamic Orientation of Nuclei [Russian translation], Mir, Moscow (1965).
9. L. N. Erofeev and B. A. Shumm, in: Radiospektroskopiya [Radio Spectroscopy] Perm', 1981, p. 54.

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