RELAXATION AND NUCLEAR MAGNETIC RESONANCE OF POLARIZED β-ACTIVE NUCLEI

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Results are presented of an investigation of the asymmetry of the radiation and NMR of polarized β-active nuclei produced by irradiating an unpolarized target by polarized thermal neutrons. The connection between the observed shape of the NMR line and the neutral line shape determined by the local fields is considered. The polarizations of the ground states of Li^8, F^{20}, A^{108}, Ag^{110} and Cu^{66} nuclei after capture of a thermal neutron are obtained. The local fields at the Li^8 nuclei in crystalline Li^7F are derived from the shape of the NMR line. When the sample is cooled to helium temperatures, an increase of asymmetry of the β emission from F^{20} nuclei and a change of the shape of the NMR curve shape in single-crystal CaF_2 were observed. It is shown that the polarized Ag^{108} and Ag^{110} β-active nuclei in AgCl crystals consist of two subsystems, one with a large relaxation time and the other with a small one.

1. INTRODUCTION

IT is known [1] that capture of polarized thermal neutrons by nuclei can yield β-active nuclei with high degree of polarization (10—50%). The polarization can be accurately monitored by determining the asymmetry of the β emission of these nuclei. This makes it possible to study a whole series of phenomena in which a change takes place in the nuclear polarization.

If a system of polarized β-active nuclei is placed in cross magnetic fields, one constant (H_0) and the other (H_1) of Larmor frequency and inducing transitions between the Zeeman sublevels of this system (the NMR method), then the populations of these levels can be equalized, i.e., the β-active nuclei can be depolarized. This will cause vanishing of the asymmetry of their β emission. This procedure was used by Connor and Tsang [2,3] to determine the g-factors of Li^8 and F^{20}.

If β-active nuclei with spin $J \geq 1$ are produced in a crystal at lattice sites with symmetry lower than cubic, then they will be acted upon, besides the external magnetic field H_0, by the inhomogeneous electric field. This makes the spacing of the Zeeman levels unequal and leads to a splitting of the NMR line of the polarized β-active nuclei: the asymmetry decrease will occur at 2J values of the radio-frequency field. From the size of the splitting it is possible to determine the interaction between the quadrupole moments of the β-active nucleus and the gradient of the electric field of the lattice.

Since the produced nuclear polarization greatly exceeds the equilibrium value determined by the Boltzmann factor, it will be decreased by various relaxation processes. Unlike the usual systems of polarized nuclei, the depolarization of the system under consideration is characterized by a number of features [1]. One of them is that the produced β-active nuclei have a gyromagnetic ratio different from that of the surrounding target-sample nuclei, and therefore the mechanism of the spin-spin exchange between the polarized system of β-active nuclei and the unpolarized system of the target nuclei will be greatly weakened. Another feature is that the compound nucleus is in an excited state after capture of the neutron, and emits γ quanta as it goes over into the β-active ground state. It acquires thereby a recoil momentum. The recoil energy amounts to hundreds of electron volts, and therefore an atom containing a β-active nucleus is knocked out of the lattice. The knocked-out atom is decelerated in the lattice, within a time 10^{-13} sec, as a result of vacancy production. According to the radiation theory [4], we can expect about ten vacancies to be produced near the place where the β-active nucleus is stopped, with a corresponding number of doubled

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atoms. These lattice damages can lead to an additional depolarization of the β-active nuclei and affect the shape of the NMR line. By observing the time variation of the β-emission asymmetry, we can determine the relaxation time and investigate its dependence on the magnetic field, on the temperature sample, on its chemical composition, and on the crystal structure.

A similar procedure was used by Wapstra and Connor [5] to investigate the asymmetry of the β decay of Li^8 and F^{20} as a function of the field H_0, and by Abov et al.[6] to study the asymmetry of the β decay of Ag^{108} and Ag^{110} as a function of the temperature. In this study we investigated polarized β-active nuclei Li^8, F^{20}, Ag^{108}, Ag^{110}, and Cu^{66} by both the NMR method and by measuring the time variation of the β-emission asymmetry, in different magnetic fields and at different temperatures.

2. THEORETICAL PREMISES OF THE EXPERIMENT

1. Emission Asymmetry of Polarized β-active Nuclei

The compound nuclei produced by capture of polarized thermal neutrons are in general also polarized. By emitting γ quanta, they go over into the β-active ground state, conserving a considerable fraction of their polarization (polarization is lost if the spin of one of the intermediate states in the γ transition is zero).

The angular distribution of the β particles emitted by the polarized nuclei is determined for allowed β transitions by the relation

$$w(\theta) = 1 + \frac{v}{c} P_n \alpha \cos\theta, \tag{1}$$

where θ is the angle between the direction of the neutron polarization P_n and the β-particle emission direction, v is the β-particle velocity, and α is an asymmetry coefficient equal to

$$\alpha = PA. \tag{2}$$

The coefficient A is determined by the nature of the β transition, P is the polarization of the β-active nucleus in the ground state when $P_n = 1$.

The experiment usually consists of observing the number of β particles emitted from the sample into a given solid angle in the direction of the neutron polarization (I_1) and in the opposite direction (I_2):

$$I_{1,2} = I_0(1 \pm \varepsilon), \tag{3}$$

where the β-decay asymmetry is

$$\varepsilon = \frac{\bar{v}}{c} P_n \alpha \Omega F. \tag{4}$$

The bar over v/c denotes averaging of the electron velocity over the β spectrum, with allowance of self-absorption by the sample.

The quantity F determines the decrease in the observed value of ϵ as a result of the back scattering of the electrons in the sample and in the surrounding materials, and Ω is a function of the solid angle in which the decay electrons are registered. Knowledge of these parameters is necessary only for the determination of the absolute value of the polarization of the β-active nuclei. From the point of view of the study of the polarization relaxation, importance attaches to the relative character of the behavior of the polarization, which is described by the experimentally observed value of the asymmetry ϵ. Therefore the determination of these parameters is not discussed here. It is described in detail in [7].

As a result of the action of various relaxation processes that decrease the polarization of the nuclei, the polarization P at the instant of time t differs from the value P_0 at the instant of formation of the β-active nuclei. The connection between them is

$$P = P_0 \exp\{-t/T_1\}, \tag{5}$$

where T_1 is the relaxation time of the system of polarized β-active nuclei.

Since the system of polarized β-active nuclei is produced in the experiment within a finite time interval in which the sample is irradiated by polarized neutrons, and since the β-active nuclei decay and become depolarized, it follows that the polarization of the entire ensemble of β-active nuclei, at an instant of time t following the end of the irradiation, is given by

$$P(t) = P_0 \frac{\lambda}{\lambda + 1/T_1} \frac{1 - \exp\{-(\lambda + 1/T_1)t_0\}}{1 - \exp\{-\lambda t_0\}} \exp\left\{-\frac{t}{T_1}\right\}, \tag{6}$$

where t_0 is the irradiation time, and λ is the β-decay constant. It is obvious that this relation is valid also for the quantity ϵ.

If the β-decay asymmetry is measured within a time Δt (starting with the instant t after the end of the irradiation), then the experimentally observed value of the asymmetry is connected with the value of ϵ_0 by the relation

$$\varepsilon(t_0, t, \Delta t) = \varepsilon_0 \frac{\lambda^2}{(\lambda + 1/T_1)^2} \frac{1 - \exp[-(\lambda + 1/T_1)t_0]}{1 - e^{-\lambda t_0}} \times \frac{1 - \exp[-(\lambda + 1/T_1)\Delta t]}{1 - e^{-\lambda \Delta t}} e^{-t/T_1}. \tag{7}$$

The same dependence will be valid also for the quantity α.

By experimentally measuring the time dependence of ϵ and by using relation (7), we can determine the relaxation time T_1 of the polarization of the β-active nuclei.

2. NMR Line Shape of Polarized β-active Nuclei

Let us obtain the dependence of the experimentally observed asymmetry ϵ on the frequency ν of the applied radio-frequency field, i.e., the NMR line shape. To this end we first obtain (in analogy with expression (5)) the time dependence of the polarization P of the β-active nuclei produced at the same instant of time when they are acted upon by two depolarizing factors - the internal relaxation process with relaxation time T_1, and a radio frequency oscillating field with amplitude $2H_1$. The total magnetic moment of these nuclei at the instant of time t after their production is

$$M_z = \mu P P_n n, \tag{8}$$

where n is the number of β-active nuclei, $n = n_0 \exp(-\lambda t)$. The rate of change of the moment M_Z with respect to time is

$$\frac{dM_z}{dt} = \left(\frac{dM_z}{dt}\right)_{P=\text{const}} + \left(\frac{dM_z}{dt}\right)_{n=\text{const}}, \tag{9}$$

where the first term of the right side describes the change of M_Z as a result of the β decay of the nuclei, and the second is due to the action of the depolarizing factors.

If W is the probability per unit time of the transition induced by the radio frequency field, then, as shown by Abragam [8], the rate of change of the magnetic moment can be represented for a two-level system of nuclei ($J = 1/2$) with $Wt \ll 1$ in the form

$$\left(\frac{dM_z}{dt}\right)_{n=\text{const}} = -\frac{M_z}{T_1} - 2WM_z, \tag{10}$$

where the first term on the right side of the equation describes the decrease of M_Z due to the internal relaxation process, and the second is due to the action of the radio-frequency field. From (8), (9), and (10) we get

$$P = P_0 \exp\{-(1/T_1 + 2W)t\}, \tag{11}$$

where P_0 is the polarization of the β active nuclei at the instant of their production.

We express the quantity W(J) for nuclei with $J \neq 1/2$ in terms of the probability of the transition $W(1/2)$ for nuclei with $J = 1/2$ [8], in the form

$$W(J) = bW(1/2) = 1/4\, b\gamma^2 H_1^2 f(\nu), \tag{12}$$

where γ is the gyromagnetic ratio, $f(\nu)$ is the normalized function of the resonance line shape, determined by the distribution of the local intracrystalline fields at the β-active nuclei [9], and the coefficient b depends on the value of the nuclear spin J and the law governing the population of the levels of the system of the polarized nuclei. In the case of a linear population law we get

$$b = \frac{B}{2J}, \quad B = \sum_{m=J}^{-(J-1)} \frac{(J+m)(J-m+1)}{2J}. \tag{12'}$$

The factor B is the result of the averaging of the probability of the induced transition $m \to m-1$ over all the transitions [10]. The factor $1/2 J$ is the result of the fact that when the populations of two levels are equalized the magnetic moment of a many-level system changes by a factor $1/2 J$. For example, for the nuclei Li^8 and F^{20} the spin is $J = 2$ and $b = 1.25$.

The dependence of the experimentally observed asymmetry ϵ on the frequency of the applied radio-frequency field is obtained, as follows from a comparison of (5) with (11), from expression (7) by replacing $1/T_1$ with $1/T_1 + 2W$:

$$\varepsilon = \varepsilon_0 \frac{\lambda^2}{(\lambda + 1/T_1 + 2W)^2} \frac{1 - \exp[-(\lambda + 1/T_1 + 2W)t_0]}{1 - e^{-\lambda t_0}} \times \frac{1 - \exp[-(\lambda + 1/T_1 + 2W)\Delta t]}{1 - e^{-\lambda \Delta t}} \exp\left[-\left(\frac{1}{T_1} + 2W\right)t\right], \tag{13}$$

Let us consider in greater detail the line shape given by expression (13). It depends on the values of the times t_0, Δt, and t. For the case of continuous irradiation of the sample by neutrons and simultaneous registration of the asymmetry of the decay electron [3], expression (13), when account is taken of (12), takes the following form (in this case $t = 0$, $\lambda t_0 \gg 1$, $\Delta t \to 0$):

$$\varepsilon' = \varepsilon_0 \lambda [\lambda + 1/T_1 + 1/2\, \gamma^2 H_1^2 b f(\nu)]^{-1}. \tag{14}$$

For the case when the irradiation and measurement cycles do not overlap in time, assuming that $t = 0$, $\lambda t_0 \gg 1$, and $\lambda \Delta t \gg 1$, we get

$$\varepsilon'' = \varepsilon_0 \lambda^2 [\lambda + 1/T_1 + 1/2\, \gamma^2 H_1^2 b f(\nu)]^{-2}. \tag{15}$$

From (14) and (15) we obtain the ratio of the asymmetry far from resonance to the asymmetry at resonance:

$$\frac{\varepsilon_\infty'}{\varepsilon_{\nu_0}'} = 1 + \frac{\gamma^2 H_1^2 b}{2\zeta(\lambda + 1/T_1)}, \tag{16}$$

$$\frac{\varepsilon_\infty''}{\varepsilon_{\nu_0}''} = \left[1 + \frac{\gamma^2 H_1^2 b}{2\zeta(\lambda + 1/T_1)}\right]^2. \tag{17}$$

If $\lambda \gg 1/T_1$, then it follows from (14)—(17) that $\epsilon_\infty' = \epsilon_\infty'' \equiv \epsilon_0$ and

$$\varepsilon_{\nu_0}'/\varepsilon_{\nu_0}'' = 1 + \gamma^2 H_1^2\, b/2\zeta\lambda. \tag{18}$$

In these expressions, $\zeta = \delta\pi$ for the case of a Lorentz shape $f(\nu) = \delta/\pi\,[\delta^2 + (\nu - \nu_0)^2]$ and $\zeta = \delta\sqrt{\pi}$ for a Gaussian shape

$$f(\nu)=\frac{1}{\delta\sqrt{\pi}}\exp\left\{-\frac{(\nu-\nu_0)^2}{\delta^2}\right\}.$$

We denote by Δ the half-width of the experimental resonance curve (ν) at half its depth. The connection between Δ and the parameters δ σ is expressed in the following manner:

1. For a Lorentz shape:

a) curve ϵ'

$$\frac{\Delta_1^2}{\delta^2}=\frac{1/2(\varepsilon_\infty'/\varepsilon_{\nu_0}'+1)-1}{1-1/2(\varepsilon_{\nu_0}'/\varepsilon_\infty'+1)}=\frac{\varepsilon_\infty'}{\varepsilon_{\nu_0}'}=1+\frac{\gamma^2H_1^2b}{2\pi\delta(\lambda+1/T_1)}, \tag{19}$$

b) curve ϵ''

$$\frac{\Delta_2^2}{\delta^2}=\frac{[1/2(\varepsilon_\infty''/\varepsilon_{\nu_0}''+1)]^{1/2}-1}{1-[1/2(\varepsilon_{\nu_0}''/\varepsilon_\infty''+1)]^{1/2}}. \tag{20}$$

2. For a Gaussian shape:

a) curve ϵ'

$$\Delta_1^2/\delta^2=\ln[1+\varepsilon_\infty'/\varepsilon_{\nu_0}'], \tag{21}$$

b) curve ϵ''

$$\frac{\Delta_2^2}{\delta^2}=\ln\left[1+\frac{[1/2(\varepsilon_\infty''/\varepsilon_{\nu_0}''+1)]^{1/2}-1}{1-[1/2(\varepsilon_{\nu_0}''/\varepsilon_\infty''+1)]^{1/2}}\right]. \tag{22}$$

It follows from (16), (17), and (18) that for the same type of nuclei, for the same sample, and for the same amplitude of H_1, the decrease of the asymmetry at resonance in the continuous regime (ϵ') is smaller than in a regime which gives the value ϵ''. For example, if $\epsilon_\infty'/\epsilon_{\nu 0}'=2$, then under the same conditions $\epsilon_\infty''/\epsilon_{\nu 0}''=4$. It is possible to increase the ratio $\epsilon_\infty'/\epsilon_{\nu 0}'$ by increasing the amplitude of the radio-frequency field H_1. If this ratio is made equal to the ratio $\epsilon_\infty''/\epsilon_{\nu 0}''$ (i.e., $\epsilon_\infty'/\epsilon_{\nu 0}'=\epsilon_\infty''/\epsilon_{\nu 0}''=\epsilon_\infty/\epsilon_{\nu 0}$), then we obtain from (19) and (20), for a Lorentz shape,

$$\frac{\Delta_1^2}{\Delta_2^2}=\frac{[1/2(\varepsilon_\infty/\varepsilon_{\nu_0}+1)]^{1/2}+1}{1+[1/2(\varepsilon_{\nu_0}/\varepsilon_\infty+1)]^{1/2}} \tag{23}$$

from which we see that the half-width Δ_1 of the ϵ' curve exceeds the half-width Δ_2 of the ϵ'' curve (for example, for $\epsilon_\infty/\epsilon_{\nu 0}=4$ we get $\Delta_1^2/\Delta_2^2=1.44$).

Thus, from formulas (18) and (23) it follows that to obtain a deeper and narrower NMR line it is more convenient to plot the $\epsilon''(\nu)$ curve.

The features of this method consist in the fact that the form of the resonance curve $\epsilon(\nu)$ depends on the decay probability λ per unit time. This is connected with the fact that $1/\lambda$ determines the effective time of action of the radio-frequency field on the polarized β-active nucleus. For example, the shorter the lifetime of the investigated nuclei (the larger λ) the smaller the depth of the dip in the resonance curve.

It follows directly from (19) that in order for the half-width of the experimental curve Δ to be close to the natural half-width δ, it is necessary to satisfy the condition

$$H_1^2<\frac{2\pi\delta(\lambda+1/T_1)}{\gamma^2b}, \tag{24}$$

i.e., it is necessary to employ sufficiently weak fields H_1. An experimental criterion for the satisfaction of the condition (24) is the relation

$$\varepsilon_\infty-\varepsilon_{\nu_0}\lesssim\varepsilon_{\nu_0}. \tag{25}$$

The function $f(\nu)$ of the resonance line shape can be obtained directly from the experimentally measured values of ϵ' and ϵ''. To this end we transform expressions (14) and (15) into

$$\frac{\gamma^2H_1^2b}{2(\lambda+1/T_1)}f(\nu)=\varkappa, \tag{26}$$

where κ takes on values

$$\varkappa=\varkappa'\equiv\varepsilon_\infty'/\varepsilon'-1 \tag{27}$$

or

$$\varkappa=\varkappa''\equiv(\varepsilon_\infty''/\varepsilon'')^{1/2}-1. \tag{28}$$

The half-width of the $\kappa(\nu)$ curve is equal to δ in the case of a Lorentz shape and to 0.835δ in the case of a Gaussian curve.

3. MEASUREMENT PROCEDURE

The experimental setup for the measurement of the β-decay asymmetry and NMR of polarized nuclei was described by us in detail earlier [7]. Figure 1 shows a block diagram of this setup. The investigated sample, placed in a cryostat in the gap of a magnet, is exposed for a time t_0 to polarized thermal neutrons obtained by reflection from a magnetized cobalt mirror. The decay electrons are counted by two counters located at 0 and 180° respectively to the direction of the neutron polarization, after an arbitrary specified time t elapsed from the blocking of the neutron beam. The counting-time duration Δt is the same for both counters. After the lapse of a certain time, which is needed for the decay of the investigated nuclei and the β-background due to the surrounding structural materials, the cycle is repeated with the direction of the neutron polarization reversed, making it possible to eliminate the instrument asymmetry. The asymmetry ϵ was calculated from the formula

$$\varepsilon=\frac{\sqrt{R}-1}{\sqrt{R}+1}\qquad R=\frac{(N_1/N_2)^{1\leftarrow2}}{(N_1/N_2)^{1\rightarrow2}}. \tag{29}$$

Here N_1 and N_2 are the β-counter readings; the arrow denotes the direction of the neutron polarization.

When the β-decay asymmetry was measured as a function of the frequency of the radio-fre-

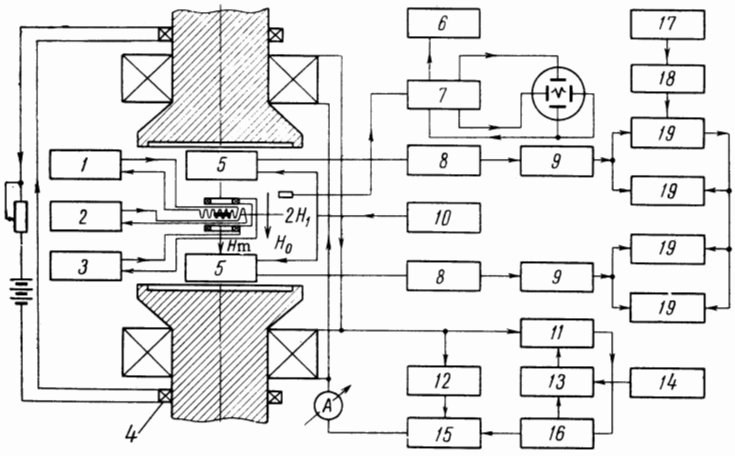

FIG. 1. Block diagram of setup for the measurement of β-asymmetry and NMR of polarized nuclei: 1–radio-frequency generator, 2–frequency meter, 3–magnetic-field modulator, 4–compensating coils, 5–electron counters, 6–heterodyne wave meter, 7–proton-resonance meter, 8–cathode follower, 9–amplifier and differential discriminator, 10–high-voltage rectifier, 11–flow-through triodes, 12–rough stabilization system, 13–comparison circuit, 14–reference voltage, 15–photomultiplier, 16–shunt, 17–neutron beam interrupter, 18–control circuit, 19–scaler.

quency field, the static magnetic field H_0 was kept constant, and the sample was subjected to the continuous action of the radio-frequency field during the entire irradiation and measurement time. To obtain the resonant value of the frequency ν_0, the static magnetic field H_0 was modulated sinusoidally at a frequency of 50 Hz, with a maximum modulation depth $\Delta H_m = 2H_m = 150$ Oe.

4. RESULTS AND DISCUSSION

1. Polarization of β-active Nuclei

The β-decay asymmetry measurements were made on Li^8, F^{20}, Ag^{108}, Ag^{110}, and Cu^{66}. At room temperature, asymmetry was observed on Li^8 in polycrystalline Li^7F and on F^{20} in single-crystal CaF_2. For the isotopes Ag^{108}, Ag^{110}, and Cu^{66}, the asymmetry in Ag^{107} Cl, Ag^{109} Cl, and CuS was observed only when the samples were cooled to helium temperature.

The maximum β-emission asymmetry values obtained in this investigation, and the corresponding values of the polarization of the β-active nuclei, calculated from formulas (2) and (4), are listed in the table. The value of the polarization for the Li^8 nuclei is close to the theoretical value calculated from the formulas of [1]. This indicates that no depolarization of the Li^8 takes place during the time of measurement at room temperature and in magnetic fields stronger than 100 Oe. It follows from (7) that the relaxation time is in this case $T_1 \gg 1/\lambda \sim 1$ sec.

Nucleus	Sample	ε, %	P, %
Li^8	Li^7F polycrystal	-6.9 ± 0.3	44
F^{20}	CaF_2 single crystal	-3.4 ± 0.2	26
Ag^{108}	$Ag^{107}Cl$ rolled single crystal	-3.4 ± 0.1	22
Ag^{110}	$Ag^{109}Cl$ rolled single crystal	-2.4 ± 0.1	16
Cu^{66}	CuS single crystal	-1.0 ± 0.1	4

For F^{20} nuclei, the maximum value of the asymmetry was obtained in a field $H_0 = 2700$ Oe at helium temperatures. In the same field, but at room temperature, the asymmetry is much lower and is equal to $\epsilon = -(1.86 \pm 0.14)\%$. Wapstra and Connor [5] have shown earlier that at room temperature, for fields $H_0 \geq 1000$ Oe, the β-decay asymmetry of F^{20} in single-crystal CaF_2 does not depend on the magnetic field. The experimental plot of the asymmetry ϵ against the field H_0 agreed well with a relaxation-time variation $T_1 = cH_0^2$, as it should if T_1 is determined by the dipole-dipole interaction. Taking these data and the present results into account, we can state that the limiting values of the asymmetry ϵ at room and helium temperatures are different. This difference can be attributed to the existence of an additional nuclear relaxation mechanism that depends strongly on the temperature, other than dipole-dipole; this may be quadrupole relaxation. A justification for this assumption may be the fact that atoms containing β-active nuclei are knocked out from the lattice at the instant when the β-active nucleus goes over from the excited state to the ground state by γ emission. When the atoms may be located stopped in the lattice at points where the electric field has a large gradient. Since F^{20} nuclei have a spin $J = 2$, they can relax as a result of the quadrupole interaction. The radiative-recoil process can in principle lead to formation of subsystems of polarized β-active nuclei with different relaxation times.

The measurement of the β-decay asymmetry of Ag^{108} and Ag^{110} in AgCl samples, as a function of the field H_0, and the study of its behavior with time, have made it possible to draw more definite conclusions regarding the existence of two sub-

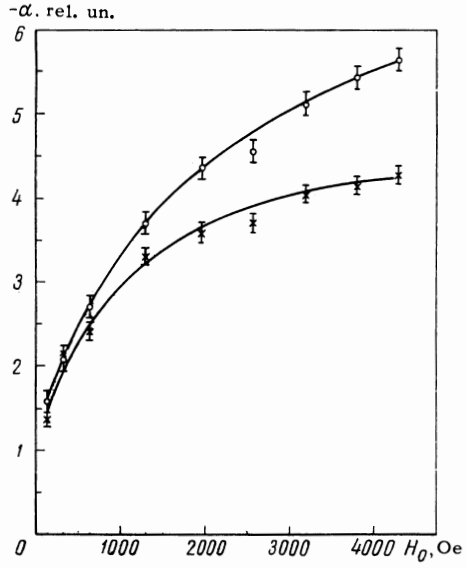

FIG. 2. Dependence of the asymmetry coefficient α on the magnetic field H_0 for $Ag^{107}Cl$ and $Ag^{109}Cl$ samples: ○–Ag^{108}, ×–Ag^{110}; T = 4.2°K.

systems of polarized β-active nuclei with different relaxation times. Figure 2 shows the dependence of the asymmetry coefficients α of these nuclei on the magnetic field H_0 on the sample. Let us assume that this dependence is determined by the field dependence of the spin-lattice relaxation time T_1. The connection between α and T_1 is given by expression (7) in which ϵ should be replaced by α, and in which we must put $t = 0$; the coefficient α_0 is equal to the value of α at $T_1 \gg 1/\lambda$, i.e., it is equal to the limiting value of α at large H_0. By determining α/α_0 from the curves of Fig. 2, we obtain with the aid of formula (7) the relaxation time T_1 for each value of the field.

These values of T_1 were used to obtain the dependence of the asymmetry ϵ on the time t for the nuclei Ag^{108} and Ag^{110} at a number of values of H_0. These are shown by the dashed curves of Figs. 3 and 4, from which it is seen that the asymmetry should decrease rapidly with time. From the experimental data shown in the same figures it follows that at values of H_0 from 140 to 4300 Oe there was no variation of the asymmetry with time. This means that the observed dependence of the asymmetry α on the field H_0 cannot be attributed to a change in the spin-lattice relaxation time T_1. From the magnitude of the experimental error we can estimate the lower limit of the relaxation time T_1 for β-active nuclei, which determine the observed asymmetry. In the case of the isotope Ag^{108} we have $T_1 \gtrsim 100$ min, and for Ag^{110} we get $T_1 \gtrsim 30$ min.

The experimentally established independence of the asymmetry of the time is evidence of the existence of a subsystem of β-active nuclei having a large relaxation time. At the same time, the field dependence of the asymmetry is evidence of a change in the number of nuclei in this subsystem. We can thus speak of the existence of at least two subsystems of β-active nuclei, one with a very large relaxation time, and the other with a small relaxation time, smaller by a factor of several times than the irradiation and measurement times (t_0 and Δt). A decrease of the irradiation and measurement time to several seconds did not lead to a change in the observed asymmetry, making it possible to establish an upper limit of relaxation time, equal to 1 sec, for the subsystem of the rapidly depolarizing β-active nuclei. If the fraction of these rapidly-relaxing β-active nuclei at a given value of the magnetic field is denoted by

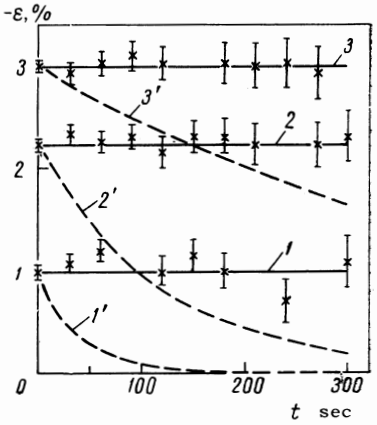

FIG. 3. Dependence of the asymmetry of β decay of Ag^{108} nuclei in $Ag^{107}Cl$ sample on the time t following the end of irradiation: 1,2,3–straight lines drawn through the experimental points for H_0 values 140, 1300, and 4300 Oe, respectively; 1',2',3'–calculated curves for the same field values.

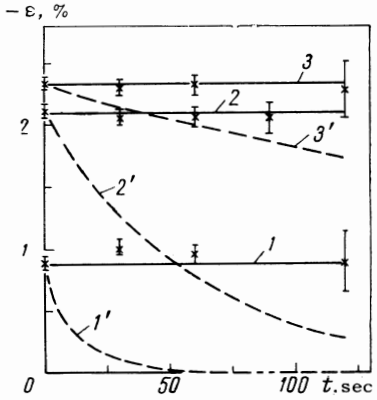

FIG. 4. Dependence of β-decay asymmetry of Ag^{110} in Ag^{109} Cl samples on the time t following the end of irradiation: 1,2,3–lines drawn through the experimental points for values of H_0 equal to 140, 1300, and 4300 Oe, respectively 1',2',3'–calculated curves for the same field values.

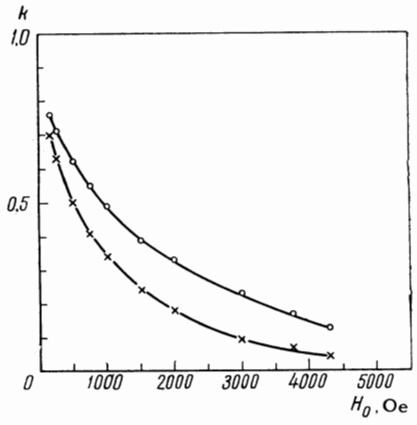

FIG. 5. Dependence of the fraction k of the rapidly-relaxing Ag^{108} and Ag^{110} nuclei in $Ag^{107}Cl$ and $Ag^{109}Cl$ samples on the magnetic field H_0: ○–Ag^{108}, ×–Ag^{110}.

$k(H_0)$, then the dependence of the asymmetry coefficient on the field is expressed as follows:

$$\alpha = \alpha_0[1 - k(H_0)]. \quad (30)$$

Figure 5 shows the fractions k of the rapidly relaxing nuclei Ag^{108} and Ag^{110} in AgCl samples, as functions of the magnetic field. The values of k were calculated by formula (30) with the aid of the curves of Fig. 2. Lowering the AgCl temperature from 4.2 to 2°K did not change the observed asymmetry. This indicates that the values of k are constant in this temperature region.

The small relaxation time ($T_1 \leq 1$ sec) for one of the subsystems of the polarized β-active nuclei can be attributed to the presence of spin-spin exchange with the unpolarized system of the target-nucleus spins, similar to the mechanism of crossing relaxation between the nuclei Li^7 and F^{19}, observed by Abragam and Proctor in the LiF crystal [8]. As follows from the NMR results presented below, the resonance lines for the nuclei Ag^{108} and Ag^{110} in AgCl should be very broad. Therefore, in spite of the large difference in the values of the g-factors of these nuclei and the remaining target nuclei [$g(Ag^{108}) \geq 10$ g (at target nuclei)], the spin exchange between them can be the result of the overlap of the lines at the fundamental frequencies. The strong dependence of the fraction of the rapidly relaxing nuclei k on the magnitude of the magnetic field also agrees with the considered spin-spin exchange mechanism.

2. Nuclear Magnetic Resonance

a) Li^8. The results of investigations of nuclear magnetic resonance of Li^8 nuclei in polycrystalline Li^7F are shown in Fig. 6. The presence of residual asymmetry of the decay electrons even at very large values of the radio-frequency field amplitude is due to the asymmetry of the β emission of the F^{20} nuclei. The NMR shape remains unchanged when the sample is cooled from room temperature to that of liquid nitrogen.

The curve drawn through the experimental points for the values $2H_1 = 0.04$ Oe was calculated from formulas (12) and (13) under the assumption that $f(\nu)$ has a Lorentz shape and that the parameters are $\delta = 1.9$ kHz and $b = b\gamma^2H_1^2/2\pi = 0.8 \times 10^3$ sec^{-2}. This value of b coincides with the calculated one within the accuracy to which H_1 was determined (20%). As noted in Item 2 of Sec. 2, formulas (12) and (13) are valid if $Wt \ll 1$. For short-lived nuclei, the time t is determined by their lifetime, i.e., $t \approx 1/\lambda$. In the case of Li^8, for $2H_1 = 0.04$ Oe and $\nu = \nu_0$, we have $W/\lambda = \overline{b}/2\delta\lambda = 0.25 \ll 1$, and the condition that Wt be small is satisfied. For $2H_1 = 0.4$ Oe we have $W/\lambda = 25 \gg 1$ and formulas (12) and (13) no longer hold. The agreement with the experimental points is obtained in this case at a lower value of δ. The half-width of the experimental curve for $2H_1 = 0.04$ Oe is $\Delta = 2.5$ kHz and is close to the

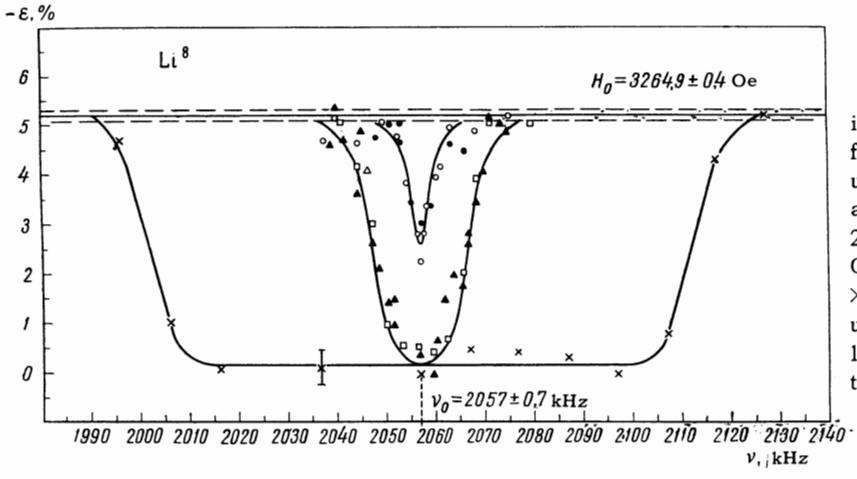

FIG. 6. Asymmetry of β decay of Li^8 nuclei in Li^7F sample vs. frequency of radio-frequency field. The experimental points were obtained under the following conditions: ○–room temperature, $2H_1 = 0.04$ Oe; ●–nitrogen temperature, $2H_1 = 0.04$ Oe; □–room temperature, $2H_1 = 0.4$ Oe; ▲–nitrogen temperature, $2H_1 = 0.4$ Oe; ×–room temperature, $2H_1 = 1.7$ Oe, field-modulation depth $\Delta H_m = 140$ Oe. The horizontal lines denote the asymmetry (with the error) in the absence of a radio-frequency field.

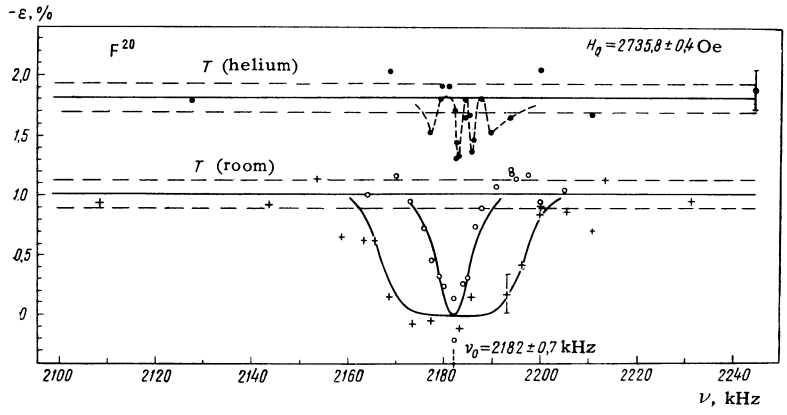

FIG. 7. Asymmetry of β decay of F^{20} nuclei in single-crystal CaF_2 vs. frequency of radio-frequency field. The experimental points were obtained under the following conditions: O—room temperature, $2H_1 = 0.04$ Oe; +—room temperature, $2H_1 = 1.6$ Oe; ●—helium temperature, $2H_1 = 0.08$ Oe. The horizontal lines denote the asymmetry (with the error) in the absence of a radio-frequency field.

natural half-width δ, in agreement with the criterion (25) for non-broadening of the line. For the curve with $2H_1 = 0.4$ Oe we have $\Delta = 10$ kHz $\gg \delta$, and at this value of the radio-frequency field amplitude the non-broadening criterion is no longer satisfied.

The value $\delta = 1.9$ kHz (± 0.4 kHz) corresponds to an interval of 6 Oe (± 1.2 Oe) of the variation of the local fields at the β-active nuclei Li^8 in polycrystalline Li^7F. Connor [2] cites for this quantity, in the case of single-crystal Li^7F, a value of 19 Oe. This value, however, was obtained from the experimental half-width Δ without allowance for the NMR line broadening. Allowance for this effect with the aid of formula (19) leads to an interval of 10 Oe for the local fields in single-crystal Li^7F. The difference between the values of the local fields obtained in the present investigation for a polycrystal and by Connor [2] for a single crystal may be connected with the anisotropy of these fields in the Li^7 lattice. The interval of the local fields for the stable nuclei Li^7 and F^{19} in single-crystal LiF is 13 Oe [8,11]. The results obtained for β-active nuclei indicate that the influence of the aforementioned radiation damage on the magnitude of the local fields at the nuclei Li^8 in Li^7F is insignificant.

b) F^{20}. The results of an investigation of the NMR of F^{20} nuclei in single-crystal CaF_2 are shown in Fig. 7. The measurements were made at two sample temperatures - room and liquid-helium.

Let us examine the results obtained at room temperature. The curve drawn through the experimental points for $2H_1 = 0.04$ Oe was calculated from formulas (12) and (13) assuming a Lorentz line shape and parameter values $\delta = 0.4$ kHz and $\overline{b} = 1.3 \times 10^3$ sec^{-2}. The obtained value of δ is low compared with the data of Tsang and Connor [3], who obtained $\delta = 0.9$ kHz at $H_0 \parallel [111]$ and $\delta = 1.5$ kHz at $H_0 \parallel [110]$. The difference is caused by the fact that we used a large-amplitude radio-frequency field ($W/\lambda = 20 \gg 1$), so that formulas (12) and (13) are not rigorously applicable[2)]. The NMR curve for $2H_1 = 1.6$ Oe has been drawn through the average value of the experimental points.

Cooling the single-crystal CaF_2 to the temperature of liquid helium produced, besides an increase in the absolute value of the asymmetry of the β decay, also a change in the NMR line shape. The dashed curve drawn through the experimental points reveals the presence of a fine structure. Such a splitting of the NMR line of the F^{20} nuclei can either be due to quadrupole interaction or to the fine structure of the magnetic field inside the crystal, both of which appear in connection with the decrease of the thermal vibrations of the atoms when the sample is cooled. For more definite conclusions concerning the nature of the splitting it is necessary to investigate the line shape by using oriented single crystals of CaF_2.

c) Ag^{108}, Ag^{110}, Cu^{66}. The magnetic moments of the nuclei Ag^{108} and Cu^{66}, determined by the magnetic-resonance method using an atomic beam [12] are: $\mu(Ag^{108}) = 4.2 \pm 0.5$ nuclear magnetons (n.m.) and $\mu(Cu^{66}) = 0.283 \pm 0.005$ n.m. However, no nuclear magnetic resonance was observed in the $Ag^{107}Cl$, CuS, and $Ag^{109}Cl$ samples for the nuclei Ag^{108} and Cu^{66} in the region of their known values of the magnetic moment, and for Ag^{110} in the range $0.073 \leq \mu \leq 10.3$ n.m., nor was resonance observed when a modulating field with depth of modulation $\Delta H_m = 150$ Oe was applied. The absence of resonance can be attributed to the strong broadening of the NMR line as a result of the interaction between the electric quadrupole moments of the nuclei Ag^{108}, Cu^{66}, and Ag^{110} with

[2)] After this article was sent to the printer, measurements with low amplitude $2H_1 = 0.004$ Oe yielded a value $\delta = 1$ kHz.

the electric-field gradients in the AgCl and CuS samples.

As shown by Abragam [8], the electric-field gradient in ionic halide crystals can be the result of the appearance of a covalent chemical bond when the atoms are displaced away from the lattice sites. In the described experiment, the displacements of the atoms can be due to the recoil of the β-active nuclei as they emit γ quanta. The occurrence of the gradient may be also due to the crystal-lattice deformation produced when the AgCl samples are rolled. The impossibility of depolarizing an appreciable fraction of the Ag^{108} and Ag^{110} by a radio frequency field signifies that the range of variation of the local electric-field gradients is very large, at least $Q\nabla E_{loc} > \mu\Delta H_m$ or $\nabla E_{loc} > \mu(150\ Oe)/Q$ (μ and Q are the magnetic and quadrupole moments of the β-active nucleus, ∇E_{loc} is the local electric field gradient). Crystalline CuS is a complex compound. The reason for the absence of NMR of the Cu^{66} in this sample is probably the same as in the case of the silver nuclei, namely a strong quadrupole line broadening due to the covalent bond.

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184