Observation of hexadecapole transitions of Al^{27} nuclei in corundum

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The allowed hexadecapole sound-induced transitions (HP) $|\Delta m| = 3$ between the levels of the magnetic resonance (MR) of Al^{27} nuclei were observed for the first time in a highly pure leucosapphire single crystal by using the method of acoustic saturation.

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The nuclei with spin $I > 3/2$ may have a fourth-order electric multipole moment. In an ionic crystal the energy of interaction of such nuclei with the surrounding medium via a hexadecapole moment is described by the Hamiltonian [1]:

$$H_{16} = \sum_{n=-4}^{4} M_{16}^{n} E_{16}^{-n} \tag{1}$$

(M_{16}^{n} are the components of the operator of a nuclear electric hexadecapole moment and E_{16}^{-n} are the parameters of the electric field of the nucleus), whose diagonal matrix elements determine the interaction of the nuclear multipole moment M_{16} with the static field of the crystal lattice and the nondiagonal matrix elements determine the nuclear hexadecapole spin–phonon coupling (NHSPC). An analysis of expression (1) shows that NHSPC is much weaker than the energies of the magnetic dipole interaction and of the electric quadrupole interaction, and, although the hexadecapole transitions (HT) $|\Delta m| = 1,2,3,4$ are allowed theoretically, in practice the single and double HT cannot be observed against the background of intensive dipole ($|\Delta m| = 1$) and quadrupole ($|\Delta m| = 1,2$) transitions. It is appropriate for this reason to study the HT $|\Delta m| = 3,4$ (the central transition $3/2 \longleftrightarrow -3/2$ is forbidden) in acoustic nuclear resonance (ANR) experiments.

Mahler *et al.* [2] attempted to detect HT. They observed sound-induced $|\Delta m| = 3$ transitions between the MR levels of In^{115} nuclei in a InAs single crystal which they attributed to the existence of HT in In^{115} nuclei ($I = 9/2$). Such interpretation of the observed saturation, however, is not correct. In analyzing the possible causes of $|\Delta m| = 3$ transitions, we must take into account the forbidden transitions due to mixing of the spin states. [3] In a number of crystals the probability of such quadrupole transitions is comparable to that of the allowed HT. In general, identification of HT in cubic crystals is associated with considerable experimental difficulties. The noncubic crystals such as Al_2O_3 are more favorable, in which the split Zeeman spectrum of Al^{27} nuclei ($I = 5/2$) allows the transitions between each pair of levels to be excited separately, and the long spin-lattice relaxation time allows experiments to be performed at room temperature.

In this paper we report the observation of an acoustic nuclear hexadecapole reso-

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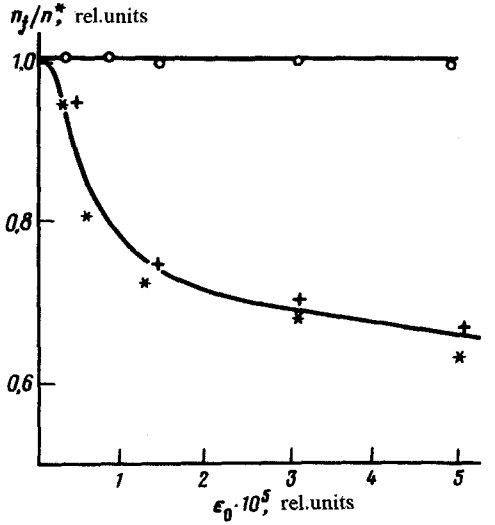

FIG. 1. Experimental dependences of the variation of the intensities of different NMR components (proportional to the difference in populations of the adjacent pairs of levels) of Al^{27} nuclei in a leucosapphire on the deformation amplitude for acoustic saturation of hexadecapole transitions $5/2 \longleftrightarrow -1/2$: +, $1/2 \longleftrightarrow -1/2$ line; O, $3/2 \longleftrightarrow 1/2$ line; *, $5/2 \longleftrightarrow 3/2$ line.

nance of Al^{27} in a Al_2O_3 single crystal of high purity (concentration of the impurities $< 10^{-5}$%). The experiment was performed at room temperature using the ANR pulsed apparatus [4] by recording the amplitude of the precession signal of Al^{27} nuclei before and after the saturating-field pulse. The sample had the shape of a $10 \times 10 \times 40$-mm^3 parallelepiped. Since the trigonal C_3 axis of the crystal was oriented in the plane of the end face, the arbitrary splitting of the lines of the MR spectrum could be determined by rotating the sample around its axis in a constant magnetic field $\mathbf{H}_0$. The characteristic quadrupole spin–lattice relaxation times T_{1k}, which were measured with accuracy of $\pm$ 5% for the satellites ($5/2 \longleftrightarrow 3/2$ and $3/2 \longleftrightarrow 1/2$) and for the central transition at maximum splitting of levels ($C_3 \| \mathbf{H}_0$), were 85, 100, and 115 seconds, respectively. The $5/2 \longleftrightarrow -1/2$ and $3/2 \longleftrightarrow -3/2$ transitions of Al^{27} nuclei were acoustically saturated at frequencies of 9.5 and 10.5 MHz by using X-cut resonance quartz plates glued to the sample's end face. The optical plane-parallel faces (laser machined) ensured the establishment in the sample of a single-mode, ultrasonic standing wave with the relative deformation $\epsilon \leqslant 5 \times 10^{-5}$. The oscillation amplitude was controlled by an electromagnetic method. [5]

The results obtained due to excitation of the $5/2 \longleftrightarrow -1/2$ transition for the case $C_3 \| \mathbf{H}_0$ are shown in Fig. 1 as dependences of relative differences in populations of different pairs of adjacent levels n_j/n^* on the deformation amplitude in the ultrasonic wave. For analogous orientation of the sample and excitation of ultrasonic $3/2 \longleftrightarrow -3/2$ transitions, we observed no variation of n_j/n^* within the limits of error of the measurements ($\pm$ 5%). The effect of different parasitic electromagnetic couplings was eliminated with great care during the experiment.

Analyzing the results by using the method described in Ref. 6, we can easily determine the probabilities of forced acoustic $|\Delta m| = 3$ transitions for different ϵ. Thus, at $\epsilon = 5 \times 10^{-5}$ we obtained $W_1 = 2 \times 10^{-3}\ \text{sec}^{-1}$ for the $5/2 \longleftrightarrow -1/2$ transition and $W_2 < 10^{-5}\ \text{sec}^{-1}$ for the $3/2 \longleftrightarrow -3/2$ transition. As is well known, since the HT $|\Delta m| = 3$ between $\pm 3/2$ levels are forbidden, the difference in the

probabilities of the investigated transitions may be due to the NHSPC. Let us estimate the efficiencies of other mechanisms of internuclear interactions, which are capable of inducing $|\Delta m| = 3$ transitions. Because of the spin–spin interactions and the interactons of nuclei with the lattice defects, the wave function Ψ_m of each level m contains an impurity of spin states with different m', which makes possible the forbidden transitions. The experimental values of the forbidden quadrupole transition probabilities, which were determined from the data for acoustic excitation of the $1/2 \longleftrightarrow -1/2$ transition, amounted to less than 10^{-5} sec^{-1} at $\epsilon = 5 \times 10^{-5}$. The observed difference in the probabilities of the induced transition witness against the inclusion of quadrupole-quadrupole interactions of the neighboring Al^{27} nuclei in the process of acoustic saturation, and the magnetic octupole transitions are unlikely, because even the acoustic dipole transitions are noticeable at the sensitivity limit of the ANR methods.[7]

Analogous investigations using a slightly dyed ruby ($10^{-3}\%$ Cr^{3+}) showed that the saturation of the $5/2 \longleftrightarrow -1/2$ transition was exactly the same as that of the $3/2 \longleftrightarrow -3/2$ transition.

Thus, the conducted investigation allowed us to identify for the first time the nuclear hexadecapole transitions and thereby to demonstrate the existence of electric hexadecapole moment in the Al^{27} nuclei.

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