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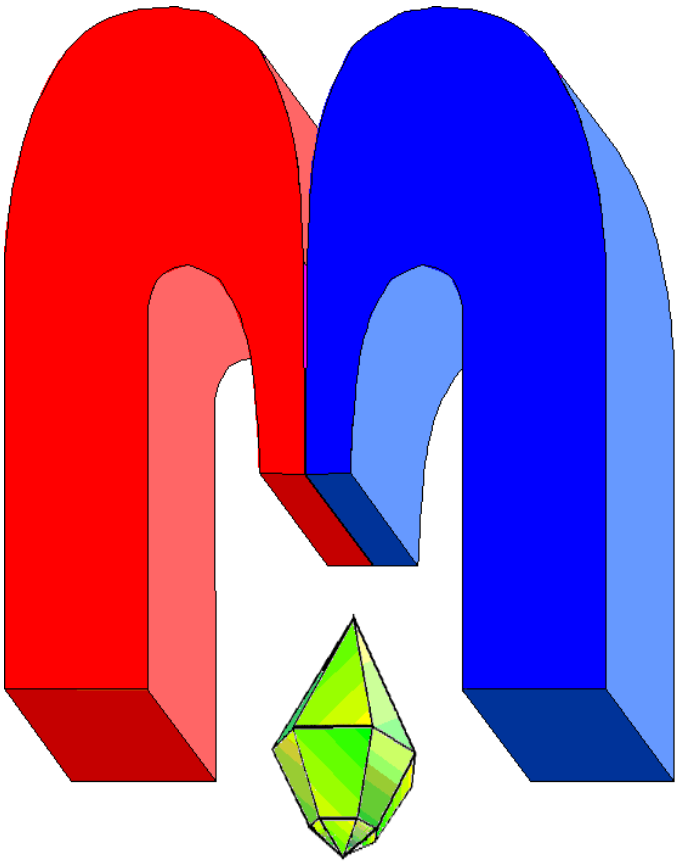

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* Address: "Magnetic Resonance in Solids. Electronic Journal", Kazan Federal University; Kremlevskaya str., 18; Kazan 420008, Russia

† In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

‡ Dedicated to Professor Boris Z. Malkin on the occasion of his 85th birthday

# Using acoustic resonance for measuring magnetostriction†

A.S. Parfishina[1,*], A.V. Dooglav[1], M.E. Menshikov[1], E.I. Baibekov[1], S.L. Korableva[1], I.V. Romanova[1], M.S. Tagirov[2]

[1]Kazan Federal University, Kazan 420008, Russia

[2]Institute of Applied Research, Tatarstan Academy of Science, Kazan, 420111, Russia

**E-mail: arina.parfishina@gmail.com*

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To measure magnetostriction in $LiTmF_4$ and $LiDyF_4$ single crystals, the acoustic resonance method was used. It is shown that the combination of capacitive dilatometry and the acoustic resonance method makes it possible to measure not only the field dependence of the crystal dimensions but also the field dependence of the sound speed.

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**Keywords:** magnetostriction, rare-earth single crystals, Van Vleck paramagnets, antiferromagnets, magnetoelastic interactions, acoustic resonance.

## 1. Introduction

The phenomenon of giant magnetostriction in compounds with rare-earth elements was discovered in 1961 in the scientific group of K.P. Belov [1, 2]. In the intermetallic compounds $ReM_2$ they studied, where Re are rare-earth ions and M are metals, the relative elongation of the samples $\Delta l/l$ reached values of $10^{-3}$ [2]. At low temperatures, the magnetostriction in such compounds was hundreds of times higher than the magnetostriction in alloys and pure metals based on iron group elements [2]. Magnetostriction was also discovered by the scientific group of S.A. Al'tshuler in dielectric single crystals of double fluorides $LiReF_4$ (Re = Tb, Ho, Er) at liquid helium temperature [3], later they also discovered giant magnetostriction, reaching values of the order of $10^{-3}$, in a single crystal of $LiTmF_4$ in fields of the order of 30 kOe [4].

Detection and measurement of magnetostriction in single-crystal samples is most often carried out using capacitive dilatometers, as well as strain gauges and holographic interferometry [5, 6].

The magnetostrictive effect is used in the creation of ultrasound generators, ultrasensitive sound receivers, and sensors for studying deformations and stresses [7]. Interest in the study of magnetostriction in rare-earth tetrafluoride single crystals is due to their potential applicability as low-temperature sensors due to the gigantic magnetostriction values in them [2–4].

Capacitive dilatometers make it possible to measure the dependence of the sample size on the magnitude of the external magnetic field. In quartz dilatometers, in a sample processed in the form of an acoustic resonator (in the form of a cylinder, the ends of which are plane-parallel), ultrasound is excited using a quartz transducer. At a frequency at which an integer number of half-waves fits along the length of the sample (with loose ends of the cylinder), a standing wave mode (acoustic resonance) appears in the sample. The resonance condition can be written as follows:

$$n\frac{\lambda}{2} = L, \quad \lambda = \frac{v}{f_n}, \tag{1}$$

whence

$$\frac{L}{v} = \frac{n}{2f_n}, \quad n = \frac{f_n}{f_{n+1} - f_n}, \tag{2}$$

†This paper is dedicated to Professor Boris Z. Malkin, who made a significant contribution to the field of magnetic radio spectroscopy in Kazan University, on the occasion of his 85th birthday.

where $L$ is the length of the sample, $v$ is the speed of sound in it, $n$ is the number of half-waves that fit along the length of the acoustic resonator, $f_n$ is the resonance frequency. The frequency corresponding to $n = 1$ is called the frequency of the fundamental tone of the resonator, frequencies corresponding to $n = 1, 2, 3, \ldots$ are called the overtone frequency. Unfortunately, the frequency $f_n$ depends not only on the size of the sample (length $L$), but also on the speed of sound, which can also depend on the strength of the magnetic field. It can be seen that decreasing the length of the resonator leads to an increase in the overtone frequency $f_n$, while decreasing the speed of sound leads to a decrease in $f_n$. A combination of measurements using a capacitive dilatometer and acoustic measurements obviously makes it possible to determine the dependence of the speed of sound in the crystal on the magnitude of the magnetic field.

The presence of quartz, of course, affects the frequency of the overtones, correcting the $L/v$ ratio:

$$\frac{L}{v} = \frac{1}{2}\left(f'_{n+1} - f'_n\right)^{-1}\left[1 + \frac{2\rho_t L_t}{\rho L}\right]^{-1} \tag{3}$$

where $f'_{n+1}$ and $f'_n$ are the overtone frequency of the composite resonator, $L_t$ and $\rho_t$ are the length and density of the quartz transducer, $\rho$ is the density of the sample.

The second way to measure the $L/v$ ratio is to use the phenomenon of magnetostriction of the crystal under study itself. If a sample with a strong dependence of size on the field is placed in an alternating magnetic field, ultrasound should be excited in the sample. An alternating magnetic field was created in the inductor of a parallel oscillatory circuit. A sample processed in the form of an acoustic resonator was placed inside the coil. When the resonant frequency of the parallel circuit coincided with the acoustic resonance frequency, a dip was observed at the top of the resonant characteristic of the parallel circuit. It is clear that this technique is no different from the technique using a composite acoustic resonator, but it eliminates the problems associated with the influence of quartz. This method made it possible to measure the field dependence of the $L/v$ ratio in all constant magnetic fields in which the crystal is subject to magnetostriction, i.e. in such fields in which the derivative of the dependence of the sample size on the field differs noticeably from zero. Unfortunately, the considered method allows one to study the temperature dependence of the $L/v$ ratio only at low temperatures and in fields in which magnetostriction has a noticeable value.

In this work, we tested the use of the acoustic resonance method to detect magnetostriction in single crystals of dielectric rare-earth tetrafluorides $LiTmF_4$ and $LiDyF_4$. The propagation velocities of ultrasonic waves along selected propagation directions are calculated using elastic constants calculated within the framework of the theory of magnetoelastic interactions developed by B.Z. Malkin [8]. The measurements were carried out in the range of magnetic fields 0–8 T at a temperature $T = 5\,\mathrm{K}$.

The studied single crystals of rare-earth tetrafluorides $LiTmF_4$ and $LiDyF_4$ are Van Vleck paramagnet and antiferromagnet, respectively. Van Vleck paramagnetism is characterized by a huge internal magnetic field which is created by the induced magnetic moment and hundreds of times greater than the external magnetic field, therefore it is also called polarization paramagnetism [4]. The $LiDyF_4$ single crystal is a dipole antiferromagnet, the Néel point is $T_N = 0.610(15)\,\mathrm{K}$ [9]. These $LiTmF_4$ and $LiDyF_4$ single crystals were grown by the Bridgman-Stockbarger method in the MRS (KFU) laboratory. The growth of single crystals of optical quality was carried out along the easy magnetization axis: in the case of $LiTmF_4$ – along the [100] axis, for $LiDyF_4$ – along the [110] axis. Both single crystals have the tetragonal structure of scheelite $CaWO_4$, space symmetry group $C_{4h}^6\,(I4_1/a)$ [10]. The search for crystallographic axes

and quality control of single crystallinity was carried out using a Bruker D8 Advanced X-ray diffractometer.

## 2. Measurement techniques and results

Static magnetostriction in single crystals of double fluorides $LiTmF_4$ and $LiDyF_4$ was measured by the acoustic resonance method. Single crystals acted as acoustic resonators in which ultrasonic vibrations were excited at resonant frequencies.

Each single crystal was placed inside the inductor of a parallel oscillating circuit and acted as an acoustic resonator, with its own quality factor significantly exceeding the quality factor of the parallel circuit. As is known, the impedance of a series oscillating circuit, which is the equivalent circuit of an acoustic resonator, at the resonant frequency is minimal and purely active. The impedance of the parallel oscillatory circuit at the resonant frequency is maximum, reaches tens of kilo-ohms, and is also purely active. When the resonant frequency of the parallel circuit coincides with the natural frequency of the acoustic resonator, the latter shunts the parallel circuit, and at the top of the resonant characteristic of the parallel circuit, a narrow dip is observed, the center of which corresponds to the resonant frequency of the acoustic resonator. In the experiment, the dependence of the resonant frequencies of the acoustic resonator on the magnitude of the external constant magnetic field was measured (Figure 1).

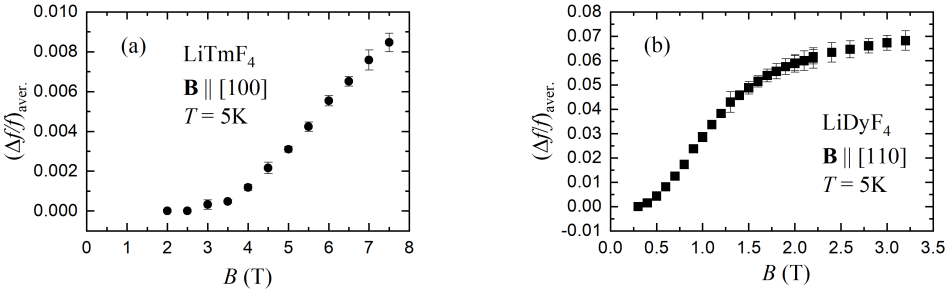


**Figure 1.** Field dependence of the relative shift of resonant frequencies, averaged over three overtones, in $LiTmF_4$ (a) and $LiDyF_4$ (b) single crystals.

## 3. Discussion

Figure 1a shows that the utilized measurement method is not not sensitive to magnetostriction when its value is too small (there are no points at $B < 2\,\mathrm{T}$). Measurements using a quartz transducer showed that in $LiTmF_4$ crystal, in the field range 0–2 T, the resonant frequency of the acoustic resonator also does not depend on the field strength. Measurements on a capacitive dilatometer show that the length of the $LiTmF_4$ single crystal changes in this range of fields [4]. We believe that the discrepancies are due to the fact that in our case, the increase in the resonant frequency due to a decrease in the length of the sample is compensated by its decrease due to a decrease in the speed of sound in the crystal.

To interpret the obtained results correctly, the field dependence of elastic constants $C_{ij}$ should be considered. Calculation of $C_{ij}$ for $LiTmF_4$ and $LiDyF_4$ single crystals was carried out with the use of parameters of electron-deformation interaction discussed previously [11]. The results are shown in Figure 2 and Figure 3 for $LiTmF_4$ and $LiDyF_4$, respectively. Previously, the attempt to take into account the temperature and magnetic field dependence of $C_{ij}$ was presented [11], but the calculated magnetic field dependencies of the elastic constants were not given explicitly.

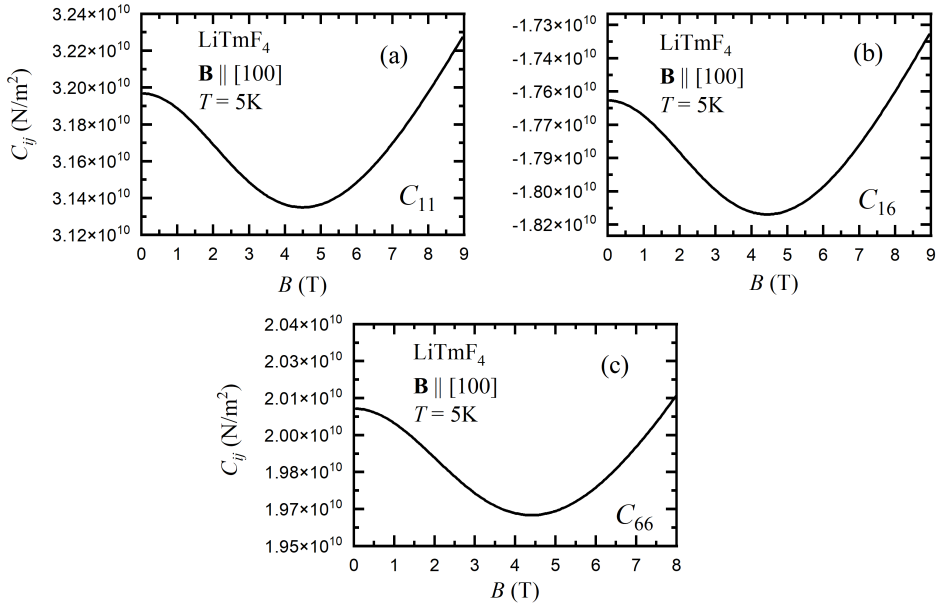


**Figure 2.** Field dependence of the elastic constants $C_{11}$ (a), $C_{16}$ (b), $C_{66}$ (c) in $LiTmF_4$ single crystal.

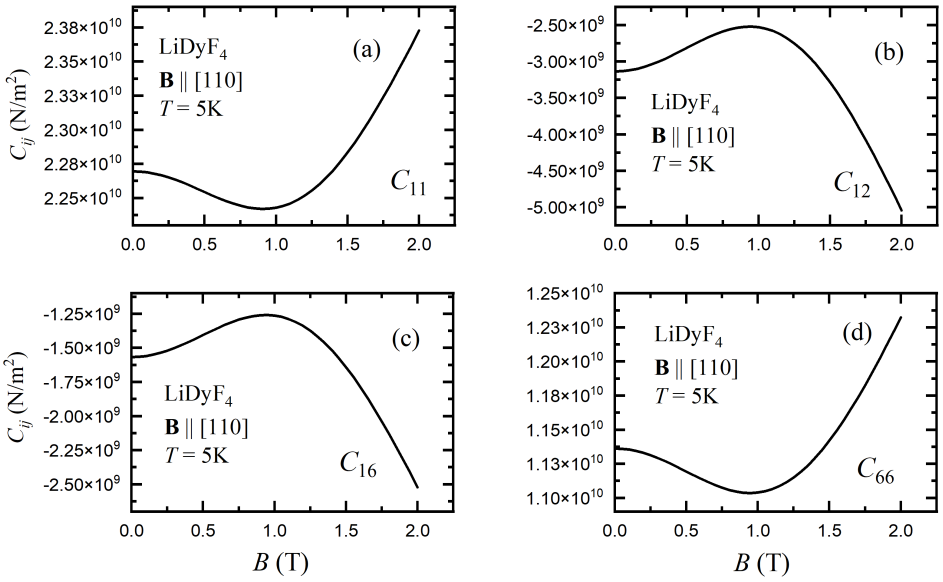


**Figure 3.** Field dependence of the elastic constants $C_{11}$ (a), $C_{12}$ (b), $C_{16}$ (c), $C_{66}$ (d) in $LiDyF_4$ single crystal.

For a longitudinal ultrasonic wave, the speed of sound is related to the elastic constants of $LiTmF_4$ crystal as follows [12]:

$$\begin{aligned} v^2_{[100]} &= \frac{1}{2\rho}\left(C_{11}+C_{66}+\sqrt{(C_{11}-C_{66})^2+4C^2_{16}}\right), \\ v^2_{[110]} &= \frac{1}{2\rho}\left(C_{11}+C_{66}+\sqrt{(C_{12}+C_{66})^2+4C^2_{16}}\right), \end{aligned} \tag{4}$$

where $\rho$ is single crystal density, $C_{ij}$ are the elastic constants.

Using the field dependence of elastic constants, we found that in the field range of 0–4 T for $LiTmF_4$, the speed of sound actually decreases with the increasing field.

## 4. Summary

Data on magnetostriction were obtained by the acoustic resonance method in single crystals of double rare-earth fluorides $LiTmF_4$ and $LiDyF_4$. In the case of a single crystal of the van Vleck paramagnet $LiTmF_4$, the magnetostriction effect is detected in magnetic fields above 2.5 T. In the antiferromagnet $LiDyF_4$, giant magnetostriction is detected using acoustic resonance already in fields above 0.3 T. The obtained curves qualitatively describe the behavior of magnetostriction in $LiTmF_4$ and $LiDyF_4$ single crystals, obtained earlier in works [4, 11, 13]. To accurately interpret the obtained results, it is necessary to take into account temperature and magnetic field dependencies of the elastic constants and consequently the change of velocity of sound in $LiTmF_4$ and $LiDyF_4$ single crystals.

In combination with the capacitive dilatometry method, the acoustic resonance method can also provide information on the dependence the speed of sound in the crystal on the magnitude of the external magnetic field and expand the understanding of internal processes in double fluoride single crystals.

## Acknowledgments

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