UDC 538.9

# LONG-TERM MEMORY AND MAGNETOACUSTIC RESPONSES IN MAGNETITE.

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**Abstract** The stimulated domain-acoustic echo DAE, possessing the long-term acoustic memory and magnetoacoustic responses were inductively excited in magnetite using a pulsed NMR technique with recording inductively the signals and studying their properties.

The comparison of the DAE intensity and magnetization dependence on the outer magnetic field showed that the long-term memory of DAE is caused by the irreversible changes of magnetization in the grains of magnetite powder due to the magnetostriction at the interaction of the acoustic wave generated by the first RF pulse with the magnetic field of the second RF pulse

*Keywords* Stimulated echo, domain-acoustic echo, long-term memory, magnetite powder, magnetoacoustic response, pulsed NMR.

#### 1. Introduction

After the excitation of a magnetostrictive sample mounted in the RF coil of a conventional pulsed NMR spectrometer by RF pulses, different magnetoacoustic responses were observed [1,2]. In [2] the domain-acoustic echo (DAE) signals were inductively generated using a pulsed NMR technique after application of three RF pulses in  $Co_{0.01}Mn_{0.05}Cu_{0.18}Ni_{0.2}Mg_{0.72}Fe_{0.6}O_4$  ferrite, europium garnet and iron borate samples at 20 MHz frequency.

The acoustic signal  $\sigma_1 = \sigma_{10} \cos(kx \cdot \omega t)$  ( $\sigma_{10}$ ,  $\omega$  and k are the amplitude, the angular frequency and the wave number of the wave, respectively) propagating in a ferrite rod in the *x*-direction after the application of the first RF pulse is recorded using the second short RF magnetic field pulse  $H_1 = H_{10} \cos(\omega t)$  of the same frequency  $\omega$ . As a result of the simultaneous action of acoustic and magnetic pulses, due to the nonlinear interaction of these oscillations with the magnetic field of the second pulse the principal contribution to which apparently comes from the magnetostriction, a stationary space-periodic magnetic structure is formed in the sample with spatially inhomogeneous magnetization component  $\Delta M \sim \cos kx$ .

This structure is a magnetic image of the acoustic signal [3,4]. The formation of this structure could be explained by the irreversible changes in magnetization. The information storage duration is practically unlimited. The information is read by the third RF or the acoustic pulse  $\sigma_2=\sigma_{20}\cos(kx-\omega t)$  as a DAE signal. In the case of DAE, the irreversible change in magnetization could be caused by the displacement of domain walls (DW) in ferrite grains [3]. In the monodomain nanosized grains, this change in magnetization is due to the change of domain magnetization caused by magnetostriction [4]. The DAE phenomenon can be used for development of the DAE processors performing integral transformations of RF signals, memory devices and delay lines [5].

The main properties of DAE formed in domains of magnetostrictive materials were qualitatively well accounted for by a simple phenomenological model [4,5]. Unfortunately, this model cannot be used for the direct quantitative comparison with the experimental results because of its simplicity. So far, polycrystalline ferrites with garnet or spinel structures have generally been used for DAE investigations [7]. Magnetite (Fe<sub>3</sub>O<sub>4</sub>), one of iron oxides, is the most magnetic of all naturally occurring minerals on the Earth. The particles of magnetite are raw materials extensively used for production of magnetic fluids, microwave absorbers, chemical sensors, and in the

information storage media [6]. The results of previous investigations of the DAE effect in magnetite [7] showed that the properties of the DAE in magnetite could not be explained by the mechanism proposed in [3] for the DAE in Ni-Cu ferrite when the irreversible change of magnetization under the action of the second RF magnetic field pulse was caused by only the displacement of DWs in ferrite grains.

In this case the magnetic structure which is formed in the sample after the application of the first pair of pulses is gradually distroyed by an increasing magnetic field and, consequently, the DAE echo decreases monotonically with the increasing magnetic field up to 50 Oe. Similar results were obtained in [2] for europium garnet powder.

In magnetite a different dependence of DAE on the outer magnetic field was observed when the intensity of the DAE at first increased until it reached a certain maximum and then gradually decreased. Besides it, the DAE intensity first behaved similar to one in europium garnet [2] but then its further increase was observed in the DAE intensity dependence on the RF pulse power.

In this work we will study the nature of the DAE in magnetite in more details.

#### 2. Experimental results and their discussion

The experimental setup using pulsed NMR spectrometer [7] was capable to produce 20 MHz RF pulses with durations and powers up to, correspondingly, 10  $\mu$ s and 100 W. The DAE and magnetoacoustic response signals were observed and studied at room temperature in magnetite samples consisting of disordered naturally occurring magnetite crystalline powders with the grain mean diameter ~ 100  $\mu$  placed in a cylindrical capsule of about 1 cm<sup>3</sup> volume. The Magnetite powder was previously thoroughly magnetically treated to remove the non-magnetic impurities.

For the magnetization measurements a vibrating sample magnetometer VSM was used.

We first studied the DAE and magnetoacoustic responses of the magnetite powder with starting mean grain size  $\sim 100~\mu$ . The signals of DAE and magnetoacoustic responses of initial powder were as is shown in Figs 1a and 1b, respectively.



**Fig. 1.** (a) The oscillogram of the DAE signal in magnetite, the time interval between the first two RF pulses:  $\tau_{12}=5 \ \mu$ s; the RF pulse duration  $\tau=5 \ \mu$ s. The oscillogram was taken at room temperature, external field 0 and 70 W RF pulse power. The total oscilloscope beam sweep duration is 100  $\mu$ s, 1 – the third RF "Read-out" pulse position marked as 1, the DAE signal position – 2. (b) oscillogram of magnetoacoustic response in magnetite under excitation by a train of RF pulses with repetition rate 1 kHz and RF pulse duration  $\tau=1.7 \ \mu$ s. The oscilloscope beam sweep duration is 100  $\mu$ s.



Fig. 2. The DAE signal dependences on RF pulse power P in magnetic at magnetic field strength (a)  $H_0=0$  and (b) on outer magnetic field  $H_0$  at P=70 W.

It was also established that the signal of DAE was practically unobservable after already 1 hour of milling and the signals of magnetoacoustic responses were also gradually disappeared (Fig. 3) along with the increase of the milling time. The magnetization measurements by the vibration magnetometer (VSM) showed a considerable increase in the coersitivity force of the magnetite powder, which could be, in our opinion, the reason for the strong reduction of DAE and magnetoacoustic response signals with milling, Fig. 4.



**Fig. 3**. Magnetoacoustic responses in the magnetite powder a) without milling, b) after 1- hour milling, c) after 5- hour milling

In Figs 4 the magnetization hysteresis curves of the investigated samples are presented.



**Fig. 4**. Magnetization hysteresis curve for the (a) initial magnetite sample; (b) 1 hour ball-milled magnetite sample and (c) 5 hour ball-milled magnetite sample.

The basic DAE properties in magnetite are different than those observed in polycrystalline  $Ni_{0.97}Cu_{0.03}Fe_2O_3$  ferrite [3] and for europium garnet in [2] and resemble the ones of more complicated behavior for iron borate [2]. It should be noted that the threshold RF power for DAE excitation in magnetite was lower as compared with other magnetostrictive materials studied by us so far. As the magnetic field increases, the DAE signal first grows, and then at the magnetic field strength H<sub>0</sub>~550 G it reaches a maximum. This result correlates qualitatively with the increase of magnetization and susceptibility in this sample, Fig.4a.

This observation allows us to conclude that the domain-echo effect in magnetite is caused by the mechanism similar to the one described in works [4,8] when irreversible changes of magnetization under the action of the second RF pulse is caused by the magnetization change in the monodomain magnetite grains due to first by domain walls displacement at lower magnetic field strength and then by the rotation of magnetization in domains at higher magnetic fields.

#### 3. Conclusion

The stimulated DAE possessing the long-term acoustic memory was inductively excited by three RF pulses in magnetite using the pulsed NMR technique with inductive recording of DAE signals.

The comparison of the DAE and magnetization dependences on the outer magnetic field showed that the long-term memory of DAE was caused by the irreversible changes in the magnetization due to first by domain walls displacement and then by domain magnetization rotation in grains due to the magnetostriction under the action of the second RF pulse.

### Acknowledgements

This work is supported by the Shota Rustaveli National Science Foundation of Georgia & Science and Technology Center in Ukraine (SRNSFG & STCU) founded targeted grant # STCU2017-31.

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Article received: 2019-11-28