[4]G. B. Scott, D. E. Lacklison, and J. L. Page, Phys. Rev. B10, 971 (1974). G. B. Scott, D. E. Lacklison, J. L. Page, and H. J. Ralf, Phys. Rev. B12, 2562 (1975). G. B. Scott and D. E. Lacklison, J. Phys. C8, 519 (1975). G. B. Scott and J. L. Page, Phys. Status Solidi B79, 203 (1977).
[5]K. W. Blazey, J. Appl. Phys. 45, 2273 (1974).
[6]G. S. Krinchik, A. P. Khrebtov, A. A. Askochenskiĭ, E. M. Speranskaya, and S. A. Belyaev, Zh. Eksp. Teor. Fiz. 72, 699 (1977) [Sov. Phys. JETP 45, 366 (1977)].
[7]A. P. Khrebtov, E. M. Speranskaya, A. A. Askochenskiĭ, and Kh. I. Turkmenov, Izv. Akad. Nauk SSSR Ser. Fiz. 42, 8 (1978).
[8]F. J. Kahn, P. S. Pezshan, and J. P. Remeika, Phys. Rev. 186, 891 (1969).
[9]R. Lindquist and A. W. Ewald, J. Opt. Soc. Am. 53, 247 (1963).
[10]R. F. Miller, L. S. Julien, and A. J. Taylor, J. Phys. D5, 2288 (1972).
[11]G. Blasse, Crystal Chemistry and Some Magnetic Properties of Mixed Metal Oxides with Spinel Structure, Proefschrift, Leyden, 1964 (Russ. Transl., Metallurgiya, 1968).
[12]N. Koshizuka, Y. Yokoyama, T. Okuda, and T. Tsushima, J. Appl. Phys. 49, 2183 (1978).
[13]G. S. Krinchik, Fizika magnitnykh yavleniĭ (Physics of Magnetic Phenomena), Izd. MGU, 1976.
[14]R. Ahrenkiel and T. Coburn, IEEE Trans. Magn. MAG-11, 1103 (1975).
[15]B. Andlauer, O. F. Schirmer, and J. Schneider, Solid State Commun. 13, 1655 (1973).
[16]B. Andlauer, R. Diehl, and M. S. Skolnick, J. Appl. Phys. 49, 2200 (1978).

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Dynamic electroacoustic echo and the recording in piezoelectric powders

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The dependence of the amplitudes and phases of two- and three-pulse electroacoustic echo on the amplitudes and phases of the applied pulses is investigated. Two methods of erasing the electroacoustic recording are proposed and used to discern the three-pulse dynamic echo in the recording. Some features of the recording and erasing processes are revealed which confirm the plastic deformation mechanism.

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INTRODUCTION

The electroacoustic echo (EAE), discovered in 1970,[1,2] manifests itself in the formation of a series of signals at the instants of time $t=(n+1)\tau$ and $t=T+n\tau$ $(n=1, 2, 3\ldots)$ upon excitation of a piezoelectric specimen by three electromagnetic radio pulses at the instants of time $t=0, \tau, T$.[3-5] The signals at $t=(n+1)\tau$ are called the two-pulse echo, and those at $t=T+n\tau$ are called the three-pulse echo. The EAE is one of the most interesting and intensely studied echo phenomena at the present time. It has been observed in a large class of different physical systems. The essential condition for the formation of the echo is the presence of nonlinearity. This nonlinearity can be an internal property of the system or can manifest itself in the interaction of the system with external fields.[6,7] Some properties of the echo signals are common to both types of nonlinearities, while others are characteristic only of one or the other. Thus, from the character of the dependence of the echo amplitude on the interval between the exciting pulses, we can distinguish the dominant role of one of the mentioned types of nonlinearity.[6,7 1)] In the case of EAE in piezoelectric powders, the study of the dependence of the echo amplitude on the interval between the pulses has led to the assumption that the dominant mechanism in echo formation is the mechanism based on the nonlinearity of the system of piezoelectric oscillators itself.[3-5]

Of special interest for investigation is the long memory of the three-pulse echo, which was first discovered in single crystals at low temperatures,[9] and later in piezoelectric powders at room temperature.[10,11] It was noted that, upon increase in the interval of time between the first and third pulses T, the amplitude of the three-pulse echo of the signal falls off rapidly with a characteristic time $T_1 \sim T_2$, reaches some definite value, and is then independent of the time of application of the third pulse. Such a dependence of the amplitude of the three-pulse echo on T is explained by the fact that the three-pulse echo is the sum of two signals: one falls exponentially to zero upon increase of T (the dynamic three-pulse echo), the other does not depend on T (the recording).[10] Thus, it is possible to record the signal for a rather long period (days, weeks). In this case, the first two exciting pulses can be called the recording pulses and the third, the reading pulse.

This paper is devoted to the study of the dependence of the amplitude and phase of the EAE on the amplitudes and phases of the exciting pulses. On the basis of the results, two methods of erasing the recordings are proposed, an experimental proof is given of the compound character of the three-pulse echo, and a method is proposed for separation of the dynamic echo and the record-

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ing. The nature of the long-term memory is piezoelectric powders is discussed in the light of the obtained data.

RESULTS AND DISCUSSION

The experiments were carried out on a nuclear quadrupole resonance coherent pulse spectrometer ISP-1, developed in the Special Design Office of the Institute of Radio Engineering and Electronics of the Academy of Sciences USSR. The range of operating frequencies is 2–20 MHz. As the object of investigation, we selected crystalline powers of cesium bromate $CsBrO_3$. All the results were obtained at room temperature.

The dependence of the amplitude of the EAE signals on the amplitudes of the applied pulses for cesium bromate powder at small amplitudes is described approximately by the relations previously obtained (see Refs. 3–5 and the references there): $A_{2\tau} = A_1 A_2$ and $A_{T+\tau} = A_1 A_2 A_3$, where A_1, A_2, A_3 are the amplitudes of the first, second and third pulses, respectively. At large amplitudes of the input pulses, significant departures from the given dependences are observed. For example, proportionality of $A_{T+\tau}$ to A_3 exists only at $A_3 \leq A_{1,2}$. Further increase in A_3 leads to a rapid decay of the signal of the three-pulse echo to zero. A similar result is obtained at T greatly exceeding the dynamic decay time of the three-pulse echo. The authors of Refs. 4 and 13 obtained similar results independently. The graphic dependence of $A_{T+\tau}$ on A_3, obtained by us, agrees well with that given in Ref. 4.

The phases of the input pulses and the EAE signals were determined relative to the phase of a standard generator. Independent regulation of the phases of the first and third pulses was provided for in the ISP-1 spectrometer. A four-pulse regime was used in the study of the dependence of the phase of the echo on the phase of the first pulse. In this case, the role of the first and second exciting pulses was played by the second and third pulses, respectively, of the four pulse regime. The time interval between the first and second of the four pulses was significantly greater than the relaxation time of the two-pulse echo T_2.

For the two-pulse echo $(n+1)\tau$ the following dependence was obtained

$$\varphi_{(n+1)\tau} = \varphi_0^{(n+1)} - n\varphi_1 + (n+1)\varphi_2, \tag{1}$$

where φ_1 and φ_2 are the phases of the first and second pulses, $\varphi_0^{(n+1)}$ is the initial phase of the two-pulse echo $(n+1)\tau$ at $\varphi_1 = \varphi_2 = 0$ $(n = 1, 2, 3, \ldots)$. The dependence of the phase of the three-pulse echo on the phases of the exciting pulses has the form

$$\varphi_{T+n\tau} = \varphi_0^{(n)} - n\varphi_1 + n\varphi_2 + \varphi_3, \tag{2}$$

where φ_3 is the phase of the third pulse, and $\varphi_0^{(n)}$ is the initial phase of the three pulse echo at $\varphi_1 = \varphi_2 = \varphi_3 = 0$. The given formulas are the result of generalization of the experimental data for $n = 1, 2, 3$ in the case of change in the phases of both the first and second pulses (the two-pulse echo) and for $n = 1$ and 2 in the case of phase changes of all three pulses (three-pulse echo). The theoretical dependence of the phases of the two- and three-pulse echos on the phase of the second pulse at $\varphi_1 = 0$ was found in Ref. 4. The experimental dependences for $n = 1$ and $\varphi_1 = 0$ was obtained in Refs. 4 and 13. Our data are confirmed by the results given in these references for special cases.

On the basis of the results stated above, we have worked out two methods of erasing the electroacoustic recording. Let an electroactoustic recording be produced in a piezoelectric sample by a pair of radio pulses with carrier frequency ν_{rec} and amplitudes $A_1 = A_2 = A_{rec}$. Repeated application of a single reading pulse with carrier frequency $\nu_{read} = \nu_{rec}$ and amplitude $A_{read} \leq A_{rec}$ leads to a nondestructive multiple reading of the recorded signal. In the dependence of the amplitude of the three pulse echo on the amplitude of the third echo, the given situation corresponds to a region of proportionality of $A_{T+\tau}$ to A_3. The first method of erasing[14] consists of applying to the sample a single erasing pulse with carrier frequency $\nu_{eras} = \nu_{rec}$, the amplitude of which A_{eras} exceeds A_{rec}. As was shown above, this leads to the destruction of the recording. The excess of A_{eras} over A_{rec} depends on the magnitude of the latter, and in the range from 100 to 2000 V/cm investigated by us, it varied within the limits 20 to 100%, increasing with increase in A_{rec}. If several signals are recorded in the sample, with different frequencies ν_{rec} (here, the difference between neighboring frequencies should exceed somewhat the width of the Fourier spectrum of the recording pulses), then the erasing (with the help of the mentioned method) of one of the signals has no effect on the others. Thus the given method allows us to make selective erasing of the signals recorded at the various frequencies.

The following evident fact lies at the base of the second erasing method[15]: as a result of the superposition of two signals, the only difference between which is a phase shift of 180°, a signal is obtained that is equal to zero. As is seen from the relation (2), the phase of the recording ν_{rec} in the absence of a reading pulse is determined by the quantity

$$\varphi_{rec} = \varphi_0^{(n)} + n(\varphi_2 - \varphi_1)_{rec} = \varphi_0^{(n)} + n\Delta\varphi_{rec}, \tag{3}$$

where $\Delta\varphi_{rec} = (\varphi_2 - \varphi_1)_{rec}$ is the phase difference between the recording signals. The initial phase $\varphi_0^{(n)}$ does not depend on the phases of the pulses and is a constant under otherwise similar conditions. The gist of the method is the superposition on the erased recording of a new recording, which differs only by the phase shift of 180°. For this, it is necessary to apply a pair of erasing pulses, choosing the phase difference between them in corresponding fashion. The remaining parameters of the erasing pair of pulses should correspond to the parameters of the pair of recording pulses. The phase difference between the erasing pulses $\Delta\varphi_{eras}$ is determined from the condition

$$\varphi_{eras} = \varphi_0^{(n)} + n\Delta\varphi_{eras} = \varphi_{rec} + \pi, \tag{4}$$

whence

$$\Delta\varphi_{eras} = \Delta\varphi_{rec} + \pi/n. \tag{5}$$

Erasing of signals with $n = 1$ and $n = 2$ was performed in the experiment. The phase difference differed by 180 and 90° from $\Delta\varphi_{rec}$, in complete agreement with the relation (5). Subsequent reading confirmed the fact that the corresponding recording was completely erased.

Several signals can be recorded at a given frequency ν_{rec}, differing in the intervals between the recording pulses. To erase one of the signals, it is necessary to choose the corresponding interval between the pulses of the erasing pair. The remaining recorded signals are preserved in this case. Moreover, this method, as the first, is selective relative to signals recorded at different frequencies. Thus, the second method allows us to use the information capacity of the sample more effectively.

The described methods of erasing an electroacoustic recording allow us to prove directly that the signal of the three-pulse echo is a sum of two signals, the dynamic echo and the static recording, and also to separate the one signal from the other.

We apply three pulses to the sample at the instants of time 0, τ and T, where $\tau < T_2$. The instant of application of the third pulse T should not exceed the decay time of the three-pulse echo T_1. The oscillogram (Fig. a) shows a series of two-pulse echo signals at instants of time that are multiples of τ after the first pulse and the three-pulse echo $T+\tau$ signal after the third pulse. Repeated application of the reading pulse confirms the existence of the recording (Fig. b). We now apply a pair of erasing pulses and a third pulse at an instant of time not exceeding the decay time of the three-pulse echo. As was shown above, the application of a pair of erasing pulses leads to erasure of the recording. Nevertheless, after the third pulse, an echo signal is observed (Fig. c) if the instant of application of the third pulse satisfies the above mentioned condition. The application of a reading pulse after a time exceeding the decay time of the three-pulse echo indicates the absence of the recording (Fig. d). The signal of the three-pulse echo in the absence of the recording falls off monotonically to zero upon increase in T, which confirms its dynamic nature. This experiment proves unambiguously that the three-pulse echo is a sum of dynamic-echo and recording signals, and also allows us to isolate one of the signals in pure form.

The study of the processes of recording and erasing revealed one important feature of the long-time memory of the EAE. The application of pulses of a definite amplitude (for example, in recording) to a sample brings the sample to a state in which recording at the same frequency by pulses with smaller amplitudes does not occur during the course of a definite time. In this case, a dynamic echo signal is formed after the reading pulse, and falls off to zero upon increase in T. We note that this effect also allows us to obtain the dynamic three-pulse echo in pure form.

In order to understand the reason for the loss in sensitivity of the sample to recording, which was also noted by the authors of Ref. 13, it is necessary to consider the mechanisms of formation of the electroacoustic recording. The basis for the so-called orientation mechanism[16-20] is the fact of the almost complete destruction of

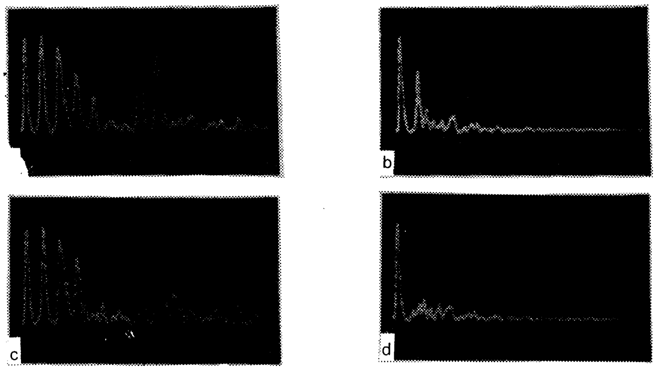

FIG. 1. Series of oscillograms illustrating an experiment on the separation of the dynamic three-pulse echo and the recording, a) Application of three pulses at the instants of time 0, τ, T leads to the generation of the two-pulse 2τ, 3τ, 4τ and the three pulse $T+\tau$ echos. b) Reading pulse, applied after several seconds, reproduces the recording. c) A pair of erasing pulses is applied. The phase of the second pulse is shifted by 180° with respect to the phase of the second pulse of case a). After the third pulse, the dynamic three-pulse echo appears. d) Reading pulse, applied within several seconds after the preceding three pulses, confirms the absence of the recording. The sample is $CsBrO_3$. The amplitudes and the carrier frequencies (9 MHz) of all the pulses are the same. $\tau = 30$ μsec.

the recording when the powder is mechanically stirred. According to this mechanism, the particles of the piezoelectric powder oscillating after the action of the first pulse are dipoles. The action of the electric field of the second pulse leads to a rotation of the axis of the dipole through an angle whose magnitude depends on the phase of the oscillations of the particles at the time of action of the second pulse. Thus, the information is fixed in the angle of rotation of the particles. Other mechanisms assume that, just as in the case of single crystals,[9] a change takes place in the internal state of the powder as a consequence of the redistribution of the electron charge,[21] of dislocations, or of point defects.[10,11] For this reason, they can be called internal. The simultaneous action of the internal and orientational mechanisms is also assumed.[4,22]

The results set forth above can be explained if we introduce the recording mechanisms recently proposed by Kessel'.[23] This mechanism is based on the phenomenon of residual plastic deformation: sign-alternating viscoplastic deformations are produced under the action of the pulses and lead to the so-called limiting state,[24] which is characteristized by residual plastic deformation of the particles. The distribution of the residual deformations depends on the phases of the oscillating particles at the time of application of the second pulse and therefore contains information on the recording. Furthermore, the limiting state is characterized by a hardening effect and, as a consequence, by an increase of the elastic modulus. Therefore, the action of pulses with smaller amplitudes no longer produces residual plastic deformations, and consequently produces no recordings. Only elastic deformations of the particles take place, leading to the appearance of the two-pulse and three-pulse dynamic echo.

CONCLUSION

1. The amplitude and phase dependences of the electroacoustic echo have been investigated and two methods of erasing the electroacoustic recording have been suggested.

2. The existence of the dynamic three-pulse echo and recording has been proved experimentally, and methods of their separation have been indicated. These methods can also be used to separate the echo signals in other systems, for example, in ferromagnetic[25] and metallic[20] powders.

3. The features of the electroacoustic recording have been made clear, and confirm the mechanism of plastic deformation.[23]

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[1)] With regard to the possibility of action of both types of nonlinearity, in accord with the experimental conditions, see Ref. 8.

[1] S. N. Popov, and N. N. Kraĭnik, Fiz. Tverd. Tela (Leningrad) **12**, 3022 (170) [Sov. Phys. Solid State **12**, 2440 (1971)].

[2] A. R. Kessel', I. A. Safin and A. N. Gol'dman, Fiz. Tverd. Tela (Leningrad) **12**, 3070 (1970) [Sov. Phys. Solid State **12**, 2488 (1971)].

[3] G. A. Smolenskiĭ, S. N. Popov, N. N. Kraĭnik, B. D. Laĭkhtman and E. A. Tarakanov, Zh. Eksp. Teor. Fiz. **72**, 1427 (1977) [Sov. Phys. JETP **45**, 749 (1977)].

[4] B. M. Berezov, V. I. Bashkov, V. D. Korepanov and V. S. Romanov, Zh. Eksp. Teor. Fiz. **73**, 257 (1977) [Sov. Phys. JETP **46**, 133 (1977)].

[5] K. Fossheim, K. Kajimura, T. G. Kazuaka, R. L. Melcher and N. S. Shiren, Phys. Rev. **B17**, 964 (1978); [Phys. Rev. Lett. **37**, 1151 (1976)].

[6] G. F. Herrmann, R. M. Hill and D. E. Kaplan, Phys. Rev. **156**, 118 (1967).

[7] R. W. Gould, Amer. J. Phys. **37**, 585 (1969).

[8] A. Billmann, Ch. Frenois, J. Joffrin, A. Levelut and S. Ziolkiewicz, J. de Phys. **34**, 453 (1973).

[9] N. S. Shiren, R. L. Melcher, D. K. Garrod and T. G. Kazyaka, Phys. Rev. Lett. **31**, 8; 9 (1973).

[10] S. N. Popov, N. N. Kraĭnik and G. A. Smolenskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. **21**, 543 (1975); [JETP Lett. **21**, 253 (1975)]; Zh. Eksp. Teor. Fiz. **69**, 974 (1975) [Sov. Phys. JETP **42**, 494 (1975)].

[11] V. M. Berezov, Ya. Ya. Asadullin, V. D. Korepanov and V. S. Romanov, Zh. Eksp. Teor. Fiz. **69**, 1675 (1975) [Sov. Phys. JETP **42**, 851 (1975)]; Ya. Ya. Asadullin, V. M. Berzon, V. D. Korepanov and V. S. Romanov, Pis'ma Zh. Eksp. Teor. Fiz. **22**, 285 (1975) [JETP Lett. **22**, 132 (1975)].

[12] A. M. Petrosyan, A. F. Volkov and Yu. N. Venevtsev, Dokl. Akad. Nauk SSSR **229**, 142 (1976).

[13] G. A. Smolenskiĭ, S. N. Popov, N. N. Kraĭnik, E. A. Tarakanov and I. A. Kuz'min, Fiz. Tverd. Tela (Leningrad) **19**, 2968 (1977) [Sov. Phys. Solid State **19**, 1738 (1977)].

[14] A. M. Petrosyan, R. I. Kukhtin, V. V. Sidorenkov, A. F. Volkov and Yu. N. Venevtsev, Inventor's Certificate No. 614466, Bulletin of Inventions (USSR) No. 25, 195 (1978).

[15] A. F. Volkov, A. M. Petrosyan, Yu. N. Venetsev and V. S. Bondarenko, Inventor's Certificate No. 624289, Bulletin of Inventions (USSR) No. 34, 167 (1978).

[16] R. L. Melcher and N. S. Shiren, Proc. IEEE Ultrasonic Symp. N. Y., 1975, p. 672.

[17] R. L. Melcher and N. S. Shiren, Phys. Rev. Lett. **36**, 888 (1976).

[18] A. A. Chaban, Pis'ma Zh. Eksp. Teor. Fiz. **23**, 389 (1976) [JETP Lett. **23**, 350 (1976)].

[19] U. Kh. Kopvillem and S. B. Leble, Ukr. Fiz. Zhurnal **21**, 954 (1976).

[20] S. Kupca, I. Maartence, H. P. Kunkel and C. W. Searle, Appl. Phys. Lett. **29**, 224 (1976).

[21] Ya. Ya. Asadullin, Z. Sh. Idiatullin and F. M. Samigullin, Pis'ma Zh. Tekh. Fiz. **3**, 289 (1977) [Sov. Tech. Phys. Lett. **3**, 117 (1977)].

[22] B. D. Laĭkhtman, Fiz. Tverd. Tela. (Leningrad) **19**, 2803 (1977) [Sov. Phys. Solid State **19**, 1642 (1977)].

[23] A. R. Kessel', Paper at the IV Intern. Conf. on Ferroelectrics, Leningrad, 1977, p. 245.

[24] V. V. Moskvitin, Plastichnost' pri peremennykh nagruzheniyakya (Plasticity under Variable Loadings), Moscow State University, 1965.

[25] N. S. Shiren and R. L. Melcher, Phys. Lett. **57A**, 377 (1976).

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