Three-pulse electroacoustic echo under conditions of acoustic resonance

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We have observed and investigated three-pulse acoustic echoes in piezoelectric crystalline powders where the crystallite dimensions satisfied the conditions of acoustic resonance. We used the usual radiospectroscopic procedure. The investigations were carried out on powdered bismuth germanate and Rochelle salt. Besides two-pulse echo signals of various types, we observed echo signals at the instant of time $T+\tau$. Following a single application of the first and second pulses and multiple repetition of the third pulse, the $T+\tau$ echo signal was observed for many days at room temperature. Results of an investigation of the properties of this echo are presented and its possible nature is discussed.

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The electroacoustic echo phenomenon, which is due to nonlinear electroacoustic effect, was observed (see^[1,2]) in investigations carried out in a two-pulse regime: radio-frequency electromagnetic pulses were applied to a piezoelectric crystal at the instants of time 0 and τ , after which a coherent acoustic response was produced at the instant 2τ and was accompanied by an electromagnetic field. Investigations were also reported^[3-7] of an electroacoustic echo following application of three electromagnetic pulses. The three-pulse echo, however, has not been sufficiently studied. Yet an investigation of its behavior is of substantial interest. The three-pulse echo was observed in the cited studies^[3-7] only at temperatures lower than 25°K.

In the present study we observed and investigated three-pulse phonon echoes in crystalline piezoelectric powders at room temperature in the entire frequency range of the spectrometer (5–70 MHz) employed in ^[1], but with a modified block for the pulse-application programs. The measurements were made on fractions of powdered bismuth germanate Bi₁₂GeO₂₀ and Rochelle salt, with particle dimensions such that the acoustic-resonance conditions were satisfied (linear dimension of the particle on the order of half the wavelength of sound) at the measurement frequency. An ampoule with the powder was placed in the capacitor of the pulse-generator tank circuit. The pulses were applied at the instants of time 0, τ , and T.

Two-pulse echoes of various types were observed: 2τ , 3τ , 4τ , $2T - 2\tau$, $2T - \tau$, 2T, and $2T + \tau$, and when the temperature was lower than room temperature also $2T - 4\tau$, $2T - 3\tau$, $2T + 2\tau$, $2T + 3\tau$, $3T - 5\tau$, $3T - 4\tau$, $3T - 3\tau$, $3T - 2\tau$, $3T - \tau$, and 3T. In addition, stimulated echo was observed at the instant $T + \tau$. This echo cannot be explained in the same way as the two-pulse echo with the aid of the parametric mechanism described by the terms fE η^2 and gE² η^2 in the expression for the crystal energy (E is the electric field, η is the strain, f and g are the coefficients of the odd and even electroacoustic effects). The echo at T + τ was observed under the condition $\tau \leq T_2$, where T_2 is the relaxation time determined from the envelope of the two-pulse echo signals at various τ . The investigations were carried out at a single amplitude $A_{1,2}$ and at equal frequencies $v_1 = v_2$ of the first and second pulses.

Figure 1 shows the amplitudes of the echo at $T + \tau$ as functions of T at constant $\tau = \tau_0$ in a regime of repeated runs of three pulses of equal amplitude and frequency. The amplitude decreases with increasing T, and the envelope can be approximately described by the exponential $\exp\{-(T - \tau_0)/T_1\}$ with a relaxation time T_1 close to T_2 . At $T - \tau_0 > T_1$, however, the echo amplitude tends not to zero but to a certain constant value, so that at $T - \tau_0 \gg T_1$ the echo practically ceases to depend on T, but a large scatter is observed in different experimental runs.

If the time $T - \tau$ of the delay of the third pulse relative to the second is kept constant, and series of threepulses are applied, with τ varying starting with τ_0 , then three-pulse echoes are observed at the instant $T + \tau$, with an envelope approximately described by the exponential $\exp(-2\tau/T_2)$ (see Fig. 2). In addition, at the instant $T + \tau_0$ it was possible to observe, as before, an echo that did not shift with changing τ . For convenience we can arbitrarily call the echo that does not depend on T "static," and the echo that varies with T and shifts and decreases in amplitude with changing τ upon application of repeated series of three pulses "dynamic."

In view of the incoherence of the applied pulses, which leads to extinction of the effect in the repeating-series regimes, we obtained enhancement of the static echo by more than one order of magnitude after a single application of the first and second pulse and repeated applications of the third pulse. In this regime the echo at T + τ had a greatly increased amplitude, which was independent, as before, of the time of delay of the third pulse for many days. In addition, in this regime the echo amplitude had high stability and a smaller scatter on going from one series of pulses to another. The results that



FIG. 1. Dependence of the amplitude of the echo at $T + \tau$ in bismuth germanate powder on T at fixed $\tau = \tau_0$ in the regime of repeated runs of three pulses.

FIG. 2. Amplitude of echo at $T + \tau$ in bismuth germanate powder against τ at a fixed delay time $T - \tau$ of the third pulse relative to the second.

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follow pertain to this regime. If the condition $\tau \ll T_2$ was satisfied, then we observed besides the $T + \tau$ echo also signals $T + 2\tau$, $T + 3\tau$, etc., i.e., the repeating third pulse reproduced, as it were, the picture of the signals produced by the first and second pulses at the instants τ , 2T, 3τ , etc.

The echo at T + τ and T \gg T₁ has a number of properties:

1) The echo amplitude A_e remains independent of T, within the limits of measurement error, for many days.

2) A_e remains unchanged after an arbitrarily large number of repetition of the third pulse at a repetition period much larger than T₂.

3) The echo effect is destroyed by changing the positions of the sample crystallites by stirring the powder relative to one another, and consequently relative to the electric-field vector of the radio-frequency pulses.

4) A_e increases with increasing amplitudes of the first, second, and third pulses (see Fig. 3).

5) The duration of the echo, measured at the base, is equal to the sum of the pulse durations, i.e., $\Delta t_e = \Delta t_1 + \Delta t_2 + \Delta t_3$.

6) When the frequency of the third pulse is changed, the amplitude of the echo goes through a sharp maximum at $\nu_3 = \nu_{1,2}$. The half-width of the spectral curve $A_e = f(\nu_3)$ is directly proportional to $1/\Delta t$ at $\Delta t = \Delta t_1 = \Delta t_2 = \Delta t_3$.

7) The frequency ν_3 corresponding to the maximum amplitude A_e at $\nu_1 = \nu_2$ increases with decreasing sample temperature (see Fig. 4) and returns to the previous value when the temperature resumes the initial value.

8) The amplitude of the echo exhibits an angular dependence when the ampoule with the sample is rotated inside the capacitor. Rotation of the ampoule through $\pm 90^{\circ}$ about its axis causes the echo to vanish. When the ampoule is returned to the initial orientation the echo reaches almost its previous value.

9) If two pulses separated by an interval τ_1 are applied to the sample, then a sequence of single pulses gives a three-pulse echo at τ_1 . If now, instead of a single pulse, a repeated pair of pulses is applied, separated by an interval $\tau_2 > \tau_1$ and having the same amplitude and frequency, then both a two-pulse echo $2\tau_2$ and three-pulse echoes are observed. The three-pulse echoes are ob-



FIG. 3. Dependence of the three-pulse echo amplitude A_e in bismuth germanate on the amplitudes $A_{1,2,3}$ of the probing pulses: curves: $1-A_e = f(A_i)$, $A_i = A_1 = A_2 = A_3$; curve $2-A_e = f(A_3)$, $A_1 = A_2 = const$.

FIG. 4. Temperature dependence of the frequency of the third pulse ν_3 , corresponding to the maximum echo amplitude A_e at T + τ .

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served at a time τ_1 both after the first and after the second pulse, and do not depend on the number of repetitions of this pair. With increasing amplitude of the paired pulses, however, the amplitude of the three-pulse echo decreases and tends to zero.

10) If a pair of pulses with τ_1 are applied once to the sample, and then a pair of pulses with τ_2 , then a multiple succeeding application of a single pulse produces two echo signals at the instants of time τ_1 and τ_2 after the single pulse. The amplitudes of the signals obtained after a successive application of pulse pairs are smaller than the echo amplitudes at τ_1 and τ_2 in the case of separate observation, when one pair of pulses is applied beforehand to the sample.

What is the nature of the investigated echo? The echo at $T + \tau$ can be observed if a dc component of the field or of the strain (or of both) is produced at the instant of action of the second pulse. This component must be modulated in space like $\cos qx$ (q is the wave vector) in accordance with the phases of the phonons from the first pulse at the instant of action of the second pulse, and must be different from zero at the instant of action of the third pulse. Then the third pulse at the instant of time T will produce phonons -q with appropriate phases, which will produce the echo signal at the instant $T + \tau$ because the oscillations are reduced to a single phase. Different interaction variants capable of producing the dc component cos qx of the field or strain are possible, including $\cos(\omega t - qx)\cos \omega t$, where the first factor can be the strain due to the first pulse or the field produced by it via the piezoeffect, and the second factor is the electric field of the second pulse. This product, depending on the participating electroacoustic effect, can describe the field and the strain. In particular, if $\cos(\omega t - qx)$ is the strain due to the first pulse, then electrostriction will produce a field $E \sim \cos qx$, and the odd electroacoustic effect will produce a strain $\eta \sim \cos qx$. The lifetime of the dc component $\cos qx$ is equal here to the duration of the second pulse (10^{-6} sec) .

A nonzero dc component can be produced after the end of the action of the second pulse by the phonons +q/2and -q/2 on account of the first and second pulses.^[6] The lifetime of this component does not exceed 10^{-3} sec under the conditions of our experiment. The amplitudes of these phonons are apparently very small and cannot explain even the dynamic component of the echo. To account for the large times of the preservation of cos qx in the case of CdS at low temperatures, a mechanism was proposed in ^[7], connected with the redistribution in space of the space charge produced upon illumination. Such a charge distribution in CdS yielded an echo at temperatures not higher than 25° K.

Observation of the echo in our case in the course of many days was evidence of prolonged preservation of the dc component cos qx resulting from the action of the second pulse. It can be concluded that the dc components of the field and of the strain, the values of which increase under conditions of acoustic resonance and which furthermore are superimposed on the resonant alternating fields and strains, cause residual microstrains and displacements of the targets. It is possible that these charge displacements and microstrains are pinned by the redistribution of the dislocations.

Attempts to produce any significant changes in the value of the echo by ionizing the air in the ampcule b_y a high-frequency discharge, and also by irradiation witl.

visible light of low intensity (including irradiation of the bismuth germanate with light of wavelength $\sim 0.44 \ \mu^{[6]}$) were not successful.

In the foregoing qualitative discussion of the effect we used the approximation of plane monochromatic waves, which are, of course, inexact under conditions of acoustic resonance. However, the proposed physical picture of the phenomenon caused by the onset of dc components of the field and of the strain components that are periodically modulated in space and reflect the phase relations of the oscillations from the first pulse at the instant of action of the second pulse, apparently reflects correctly the gist of the phenomenon. This is confirmed, for example, by the vanishing of the echo signal when the powder is stirred and when the ampoule with the powder is rotated through $\pm 90^{\circ}$, and also by the increase of the carrier frequency of the third pulse, corresponding to the maximum echo amplitude, with decreasing temperature. It appears that the latter is due to a change in the spatial period of the dc component on account of the thermal expansion, and with the change of the speed of sound with change in temperature.

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