Anomalous relaxation of stimulated echo in piezoelectric crystals

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Stimulated (three-pulse) echo was observed in a number of piezoelectric crystals, with an amplitude that decreased non-exponentially; the maximum time constant was on the order of dozens of minutes and more. Cumulative enhancement of the echo was observed when a series of pulse pairs was applied beforehand.

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We report here new results on the observation of anomalous echo phenomena in piezoelectric crystals; these phenomena were previously considered in[1-5]. We have investigated a number of ferroelectric and non-ferroelectric crystals (KDP, ADP, $LiNbO_3$, NH_4Cl, $Bi_{12}GeO_{20}$, $Bi_{12}SiO_{20}$, Rochelle salt, etc.), and in some of them the echo was observed for the first time.

To observe the effect, the samples were placed between parallel plates of a capacitor, to which a series of two or three pulses of an alternating electromagnetic field was applied. The pulse carrier frequency was the same for all pulses, $\omega/2\pi = 12.5$ or 25 MHz. The echo signals were produced only when the sample was ground into a powder; there was no echo in single crystals or in coarse-grain powders, a fact noted also in[1,2]. The investigations were carried out in the interval from room temperature to that of liquid nitrogen.

When a pair of pulses of duration $\Delta t_{1,2} \approx 2$ to 3 μsec with interval τ between them is applied, an echo is produced at the instant 2τ after the first pulse. The falloff of the two-pulse echo signal is described by a characteristic time T_2 which is different for different substances and varies with temperature. The value of T_2 (from dozens to hundreds of microseconds) and its temperature and frequency dependences indicate that this echo is due to excitation of acoustic oscillations in the piezocrystal, T_2 characterizes the irreversible damping of these oscillations.

Application of a third pulse delayed a time $\tau_1 > \tau$ relative to the first leads to the formation of stimulated echo (SE) at the instant of time $\tau_1 + \tau$. Experiments show that no exponential dependence of the SE amplitude on τ and τ_1 in the form $\exp[-2\tau/T_2 - \tau_1/T_1]$ (T_1 is a second characteristic time), is obtained with respect to τ_1. Just as in[5], the nonexponential decrease of the signal points to the existence of not one value of T_1, but of a certain distribution of its values, from fractions of a second to several dozen minutes and more. A typical variation of the SE amplitude with time is shown in Fig. 1. With decreasing temperature, the long component T_1 increases, whereas the echo amplitude decreases monotonically.

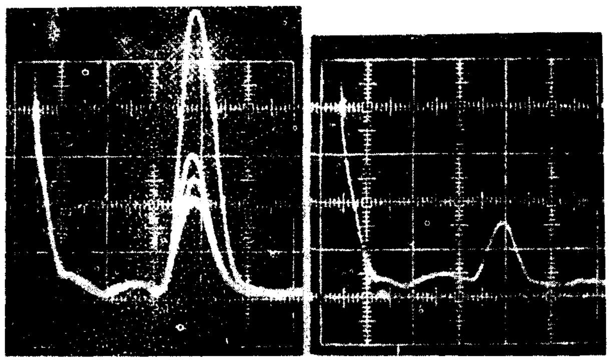

FIG. 1. Decrease of stimulated-echo amplitude with time: a) Reading downward: $\tau_1 = 0.001$, 1, 10, and 100 sec; b) $\tau_1 = 1$ hour. The gain of the receiver is the same for all oscillograms. The sample is powdered Rochelle salt with $d_{av} = 95$ μ, $\omega/2\pi = 12.5$ MHz, $\tau = 20$ μsec, $t = 20\,^\circ$C.

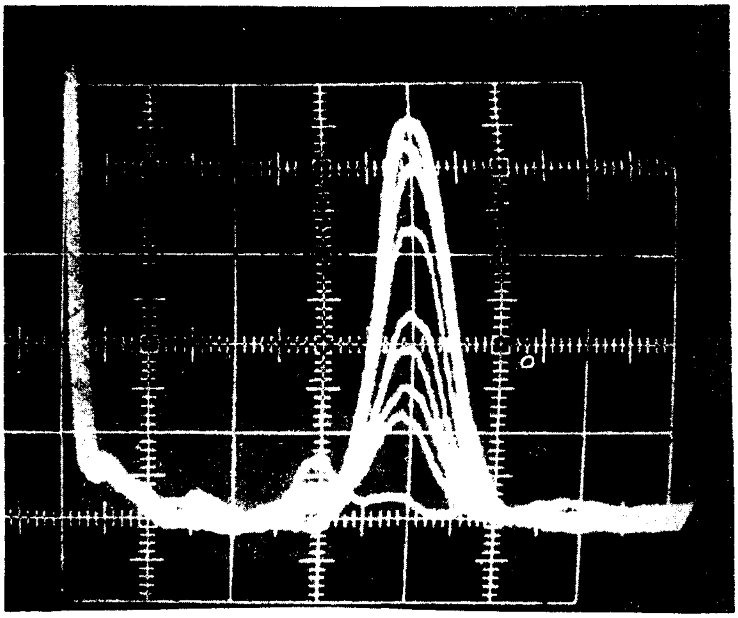

FIG. 2. Multiple exposure of SE signal as the number of exciting pairs is increased from 3 to 2×10^4, the sample and the conditions are the same as in Fig. 1.

We have observed an interesting effect of cumulative enhancement of the SE: if a series of pairs of pulses is applied instead of one pair, followed by a single third pulse, then the SE amplitude is several times larger than that produced by a single pair (Fig. 2). The enhancement depends on the pair repetition frequency and on the accumulation time. At a given pair repetition frequency, the echo amplitude becomes saturated after a definite accumulation time. This time depends on the duration and power of the pulses. It must be emphasized that in contrast to[5], the observed two- and three-pulse echo signals, and also the enhancement ef- effect, take place without any prior illumination of the investigated samples. Storing the sample for two weeks in darkness exerted no noticeable influence on the behavior of the signals.

An explanation of the SE as being due solely to phonon excitation must be rejected because of the presence of the anomalously long times T_1. It appears that this effect is due to the formation of a diffraction grating made up of electrons in "traps". [5,6] The interaction of the alternating field of the pulse witn the sound field produced by it in the sample, as well as with the sound field of the preceding pulse, leads to an interference pattern that is stationary in time and is periodic in space, with the period of the sound wave. Therefore the electrons at the maximum of the stationary-field intensity tunnel from the shallow donor level and become spatially distributed over the "traps"; a diffraction grating is then produced. The action of the next (third) pulse on the grating leads to the formation of oppositely directed sound waves, and this gives rise to the stimulated echo. In light of the foregoing, the enhancement of the SE is due to the increased contrast of the diffraction pattern under the action of the aggregate of pulse pairs. The times T_1 are determined here by the depth of the potential wells of the "traps," and the decrease of the echo amplitude with decreasing temperature is connected with the Boltzmann distribution of the electrons over the traps.

Two types of mechanism can be effective in the formation of a two-pulse echo: 1) pulse 1 produces a sound wave in the sample, and pulse 2 excites an opposing wave via some four-phonon mechanism[7]; 2) the interaction between the electric field of pulse 1 with its own sound field produces a diffraction pattern, and pulse 2 is scattered by this grating. It can be assumed that the long times, $T_2 \sim 100$ μsec and more, is due to the second mechanism.

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