

G. A. Smolenskiĭ, *The electroacoustic phonon echo*. The nonlinear effects and the interaction of "subsystems" or quasiparticles are interesting aspects of the study of solids.<sup>1</sup> Research of this type has led to the discovery of the electroacoustic phonon echo in piezoelectrics and ferroelectrics.<sup>2,3</sup>

In this effect, the application of pulses of an alternating electric field to piezoelectric samples in the form of single crystals or powders results in the appearance, at certain times, of other pulses of an alternating electric field (the echo pulses), which accompany acoustic vibrations. Piezoelectric samples subjected to two electromagnetic pulses at times  $2\tau$ ,  $3\tau$ , .... These signals constitute the two-pulse echo. If, instead, three pulses are applied to the sample, at times 0,  $\tau$ , and  $T$ , then echo signals are again emitted at  $T + \tau$ ,  $T + 2\tau$ , .... The signals at  $T + \tau$  and  $T + 2\tau$  constitute the three-pulse echo. The first and second of these pulses are the "writing" pulses, while the third is the "readout" pulse.

For the two-pulse echo to be observed, the time  $\tau$  must be smaller than the acoustic damping time  $T_2$ ; otherwise the acoustic vibrations excited in the sample by the external pulses will not interact. It would seem that  $T$  should also be smaller than  $T_2$ , but it has been found that the three-pulse echo can be observed under the condition  $T > T_2$  and even under the condition  $T \gg T_2$ . Powdered samples have revealed a long-term memory at room temperature, lasting for a matter of months.<sup>4,5</sup>

The echo is studied in two basic modes: the traveling-wave mode (with both volume and surface waves) and the mode with standing acoustic waves. In the resonant mode (standing waves) a large number ( $\sim 10^6$ ) of crystal grains are used, with dimensions of the order of half the acoustic wavelength. The echo is observed by acoustic methods or by pulsed rf spectrometry.

The appearance of this electroacoustic echo consists of the following basic steps (the same is true for echoes of other types): 1) oscillations—in the present case, acoustic—are excited; 2) these oscillations go out of

phase; 3) the time evolution of the phases is inverted (in the traveling-wave case this is an "inversion" of the wave vector; 4) the oscillations are brought into phase (the creation of the echo).

The appearance of the echo in the traveling-wave mode in single crystals can be explained as follows. The first rf pulse excites a packet of elastic waves via the piezoelectric effect, and these waves propagate away from the surface into the crystal. The displacement in this packet is

$$u_1 = C_1 e^{i(kx - \omega t)}, \quad (1)$$

where  $k$  is the wave vector and  $\omega$  is the frequency.

This packet is reflected at the ends of the crystal and travels back and forth through the crystal until it is damped. The original packet goes out of phase because of the diffraction divergence of the beam, elastic scattering of the acoustic waves by crystal inhomogeneities, and the surface roughness at the reflecting ends.

If a second rf pulse with the same frequency  $\omega$  is applied at time  $\tau$ , this pulse produces an electric field

$$E = E_2 e^{i\omega(t - \tau)} \quad (2)$$

in the crystal. The nonlinear interaction of this field with a traveling acoustic wave, which can be described, for example, by a term  $u_1^2 E^2$  in the expansion of the free energy (see below), produces a new acoustic wave in the crystal:

$$u_2 \sim u_1 E^2 \sim C_1^* E_2^2 e^{-i[kx - \omega(t - 2\tau)]} \quad (3)$$

(as usual, the asterisk denotes the complex conjugate). This new wave propagates in the direction opposite the original wave, and it can be seen from (3) that a new packet of elastic waves is produced and creates an echo signal at  $t = 2\tau$ . In the electric field of the second rf pulse, each harmonic making up the packet  $u_1$  excites a harmonic with the same phase (at the time at which the second pulse is applied) but with the opposite propagation direction. This secondary harmonic travels the same path as the original harmonic, and it undergoes the same scattering processes, but in the opposite or-

der. The net result is that the phase differences between the harmonics of the packet  $u_2$  are cancelled.

The expansion of the free energy in powers of the electric field  $E$  and the strain  $u$  can be written schematically

$$F \sim cu^2 + c^{(3)}u^3 + c^{(4)}u^4 - eE^2 + e^{(3)}E^3 + f'E^4 + eEu + dE^2u + fEu^2 + gE^2u^2 + gEu^3 + hEu^3 + \dots \quad (4)$$

It is easy to see that essentially all the terms in this expansion which describe nonlinear terms, by which we mean terms of order higher than the second, contribute to the echo.<sup>6,7</sup>

It follows from this description that the echo effect can be observed not only in piezoelectrics and ferroelectrics, i.e., in crystals which lack a symmetry center, but also in other crystals—in general, all crystals and solids. For observation of the echo in a crystal with a symmetry center, external transducers must be used to excite and transform the elastic waves. The phonon electroacoustic echo is thus a universal effect.

It is also extremely interesting to study the echo effect in fine-grain powders. For echo observation in powders, an important consideration is the condition for an acoustic resonance, i.e., the condition that the pulse-modulated frequency be approximately equal to the oscillation frequencies of the powder grains. In this case it is obviously possible to maximize the oscillation amplitudes for given electric fields. The two-pulse echo is an effect of the same physical nature in single crystals and in powders; the differences lie in the mechanism by which the various waves go out of phase and come back into phase. In single crystals, these phasing processes are due to elastic scattering of acoustic waves, while in powders they are due to the differences in the resonant frequencies for mechanical vibrations of the individual grains. Furthermore, powders are distinguished from single crystals by the fact that the most important nonlinearities are the elastic nonlinearities.

The mechanism for the formation of the three-pulse echo is slightly different. For observations in the traveling-wave mode in single crystals, the nonlinear interaction of the oscillations excited by the first two pulses leads not only to new oscillations, which produce the two-pulse echo, but also to a strain which is constant over time but inhomogeneous over space:

$$u_c \sim u_1 E = C_1 e^{i(kx - \omega t)} E_2^* e^{i\omega(t - \tau)} \sim C_1 E_2^* e^{i(kx - \omega t)}. \quad (5)$$

These spatially inhomogeneous stationary components contain information on the phases of the oscillations excited by the first and second pulses. It can be seen from (5) that these components are periodic in space with a period equal to the acoustic wavelength. The third pulse,  $u_3 = C_3 e^{-i[kx - \omega(t - T)]}$ , acting on this inhomogeneous crystal, excites oscillations with the same phase relations which prevailed at the time of the second pulse. Accordingly, the phases of these oscillations, as in the case after the second pulse, become equal at a time  $\tau$  after the application of the third pulse. In other words, at the time  $T + \tau$  the three-pulse echo appears:

$$u_e \sim u_c u_3 = C_1 E_2^* C_3 e^{-i\omega[t - (T + \tau)]}. \quad (6)$$

The inhomogeneous strain or the field in the crystal redistributes the defects and charges (the lattice is "decorated" by the defects). This inhomogeneous distribution of defects and charges can be preserved for a long time (the relaxation time  $T_1$ ), even after the oscillations which produced this distribution have been damped out. In this manner, an acoustic hologram is recorded. Under certain conditions this hologram can be read out repeatedly (an important consideration for practical applications).

In powders, a macroscopic orientational mechanism can play a definite role in creating the memory. This mechanism is due to a reorientation of the particles by the second pulse.<sup>8,9</sup>

At present, the electroacoustic echo and memory have been studied in more than 50 materials, over a broad frequency range, from 1 to 10 GHz, and over a temperature range from liquid-helium temperature to 300–400 °C. This research has established not only the mechanisms for the formation of the echo and of the memory but also the basic properties of these effects.

The electroacoustic echo has opened up a new field in the acoustics of solids. This effect is of scientific interest in its own right, and it also holds promise for research on the physics of nonlinear effects, including electroacoustic effects, the damping of elastic waves (especially at microwave frequencies), and phase transitions, and for study of the properties of materials in a finely dispersed state, lattice defects, etc.

The echo effect can be used to develop devices for information storage and processing in several technical applications: convolution, correlation, and Fourier-transformation devices; memory devices; controlled delay lines; devices for coherent and incoherent storage of rf pulses with linear and nonlinear intrafrequency modulation; and devices for time-inverting signals. Since echo devices are broad-band devices, their information storage capacity can be two or three orders of magnitude better than that of other acoustoelectric devices.

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