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Exponential growth of backward-wave phonon echoes

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A recent paper by Gaumond and Jacobsen analyzed the process of backward-wave, phonon-echo generation in an insulator in terms of solutions of Mathieu's equation. We discuss the experimental conditions under which the predictions of their analysis would be expected to hold, and we report the observation of exponential growth of the echo amplitudes in $LiNbO_3$.

Backward-wave phonon echoes have been observed in a number of different materials and under a variety of different conditions.[1] Nevertheless, numerous uncertainties still exist regarding details of the microscopic mechanisms responsible for the underlying parametric interactions. In some materials it is likely that the interaction is due to the intrinsic nonlinearity of the pure crystalline state.[2] In many samples, however, the coupling between strain and electric field is believed to arise through the agency of an impurity or a structural defect in the host material.[3]

A major problem that arises in attempting to separate the different effects is that it is difficult to carry out experiments in any but the small signal regime. In this limit, regardless of the detailed mechanism, the amplitude of the backward-wave echo u_B, has the same simple dependence on the amplitude of the pump electric field, E_P. For example, when the pump frequency is twice the frequency of the forward traveling acoustic wave, as in the experiments described below, the small signal theory predicts that u_B should be directly proportional to E_P, since this is the first term in the series expansion of several different mathematical functions.[1] Another problem has been the absence, until recently, of a general theory of the parametric interaction through intrinsic crystal nonlinearity, as opposed to the perturbation approach that gives a simple E_P dependence.

Work by Lu, Fedders, and Burgess[4] and by Gaumond and Jacobsen,[5] however, has shown that the basic equations of motion describing the coupled waves can be transformed into Mathieu-Hill equations for the Fourier amplitudes. Under some circumstances, solutions of these equations can be found for a pump field of arbitrary magnitude. A number of interesting results follow, and in an attempt to explore these features experimentally, we have been working in the regime of large intrinsic nonlinearity (using lithium niobate and lithium tantalate) high pump frequency (34 GHz) and high pump power (1–10 kW). We have indeed observed the exponential growth predicted by Gaumond and Jacobsen, and have been able to fit the data to the appropriate solution of the Mathieu equation. Before describing the results, we shall first summarize the general theory as it applies to our experimental arrangements.

The theory of parametric echo generation through intrinsic nonlinearities starts from an expansion of the free energy of the lattice V, as a Taylor series in electric field E, and strain S,

$$V = CS^2/2 + C_{NL}S^3/3! - \epsilon E^2/2 - \chi E^3/3! - eES - bE^2S + fES^2/2 + gE^2S^2/2 + hES^3/3! + \cdots \quad (1)$$

The physical significance of the different tensor coefficients is explained by Fossheim and Holt.[1] Using constitutive expressions which relate V to the elastic stress and electric displacement, together with Gauss's law and the lattice equation of motion, it is a straightforward matter to show that the lattice displacement ξ due to the acoustic traveling waves (the signal and the idler) is related to the electric field pump, through the equation

$$\rho \frac{\partial^2 \xi}{\partial t^2} = (C_0 + C_1 E_P + C_2 E_P^2 + \cdots) \frac{\partial^2 \xi}{\partial x^2} \ . \quad (2)$$

For simplicity, propagation is restricted to one dimension. The coefficients C_0, C_1, and C_2 are given by

$$C_0 = C + e^2/\epsilon \ , \quad (3)$$

$$C_1 = f + 4eb/\epsilon - e^2\chi/\epsilon^2 \ , \quad (4)$$

$$C_2 = g + 4b^2/\epsilon - 4eb\chi/\epsilon^2 + e^2\chi^2/\epsilon^3 \ . \quad (5)$$

The usual way of treating (2) is to assume that the idler wave is much smaller than the signal, and that the pump electric field is not depleted. Fedders and Lu[6] for ω-ω echoes, Economou and Spector[7] for ω-2ω echoes, and Bajak[8] for a less restrictive relationship between acoustic and microwave frequencies, have all adopted this approach. Economou and Spector[9] have also calculated the depletion of the pump on the assumption that it is an independent process. However, a more general treatment may be obtained by expanding the displacement ξ as a Fourier sum of spatial modes of wave vector k and time-dependent amplitude $A_k(t)$. Thus,

$$\xi = \sum_k A_k(t) \exp ikx + \text{c.c.} \quad (6)$$

If the pump electric field is described by $E_P = E_0 \sin\omega_0 t$ —that is, with no spatial variation of the amplitude over the interaction volume—then substitution into (6) leads to a series of equations, each of the Mathieu-Hill form

$$\frac{d^2A_k(z)}{dz^2} + (\alpha + \beta\cos 2z + \gamma\cos 4z)A_k(z) = 0 \ , \quad (7)$$

where

$$\alpha = 4(\omega/\omega_0)^2(1 + E_0^2 C_2/2C_0) \ , \quad (8)$$

$$\beta = 4(\omega/\omega_0)^2 E_0 C_1/C_0 \ , \quad (9)$$

$$\gamma = 4(\omega/\omega_0)^2 E_0^2 C_2/2C_0 \ , \quad (10)$$

$$z = \omega_0 t/2 \ , \quad (11)$$

and

$$\omega = (C_0/\rho)^{1/2} k \quad (12)$$

(ρ is the density of the material).

Since Eq. (7) cannot be exactly solved in its most general form, a simplification must be made in order to allow approximate solutions to be obtained for particular experimental circumstances. It is necessary to consider separately the situations when the coupling in Eq. (2) is either linear in E_P (so that $C_2=0$) or quadratic in $E_P (C_1=0)$. For both cases (7) then reduces to the canonical form of Mathieu's equation,

$$\left[\frac{d^2}{dz^2}+(a-2q\cos 2z)\right]A_k(z)=0 \ . \tag{13}$$

For linear coupling, $a=4(\omega/\omega_0)^2$ and

$$2q=-4(\omega/\omega_0)^2 E_0 C_1/C_0 \ ,$$

while for quadratic coupling

$$a=(\omega/\omega_0)^2(1+E_0^2 C_2/2C_0)$$

and

$$2q=-(\omega/\omega_0)^2 E_0 C_2/2C_0 \ ,$$

but with z now taking the value $\omega_0 t$. The solutions of Mathieu's equation have been widely studied[10] and are characterized in terms of the parameters a and $2q$. If these are treated as variables, the space defined by a and $2q$ divides into regions in which the solutions of (13) are either stable, when $A_k(z)$ remains bounded or tends to zero as $z\to\infty$, or are unstable so that $A_k(z)$ tends to ∞ as $z\to\infty$. On the boundaries between the stable and unstable regions, the solutions are periodic in π or 2π (Fig. 1).

In our experiments, the ratio of signal-to-pump frequencies was either 1 or $\frac{1}{2}$, since the echo (idler) had to be at the same frequency as the signal for it to be detected. This constraint restricts the parameter a to values near 1 and 4. An ω-ω echo can result from linear coupling with $a\sim 4$ and quadratic coupling with $a\sim 1$, whereas the ω-2ω echo can only result from linear coupling with $a\sim 1$. From Fig. 1 we see that at these values of a, the solutions are all unstable when $2q$ is small. However, when the map is replotted in terms of the experimentally significant parameters, coupling strength, and frequency, the second term in the expression for a for quadratic coupling, which corresponds to a shift in

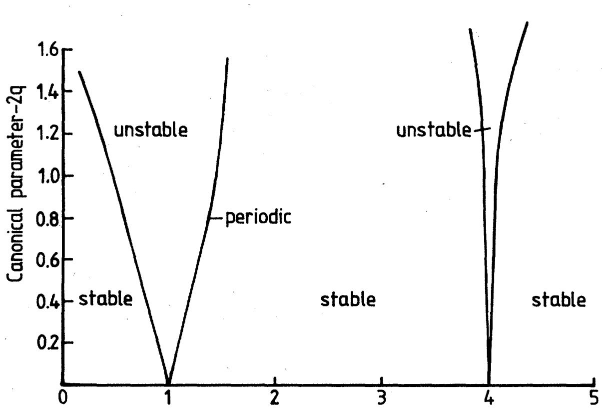

FIG. 1. The behavior of the solutions of Mathieu's equation, characterized in terms of the parameters a and $2q$.

to a shift in the velocity of the acoustic waves, causes the solutions to cross the boundary into the stable region.

Based on series expansion solutions described by Whittaker and Watson,[11] Gaumond and Jacobsen derived the following results that are relevant to our experiments:

(a) for $\omega/\omega_0=1$ with linear coupling, the amplitude of the backward wave is given by

$$u_B(T)=u_F(0)\sinh\left[\frac{\sqrt{15}}{12}\left(\frac{c_1}{c_0}\right)^2 E_0^2\omega_0 T\right] \ , \tag{14a}$$

(b) for $\omega/\omega_0=1$ with quadratic coupling,

$$u_B(T)=u_F(0)\sin\left(\frac{\sqrt{3}}{4}\frac{C_2}{2C_0}E_0^2\omega_0 T\right) \ , \tag{14b}$$

(c) for $\omega/\omega_0=1/2$ with linear coupling,

$$u_B(T)=u_F(0)\sinh\left(\frac{1}{4}\frac{C_1}{C_0}E_0\omega_0 T\right) \ . \tag{14c}$$

In the derivation of these results, it has been assumed that the spatially uniform electric field is switched on at $t=0$ when only the forward wave with amplitude $u_F(0)$ is present and is switched off at $t=T$ when both the forward and backward waves have developed. Similar expressions apply to the forward wave but with hyperbolic sine replaced by hyperbolic cosine and sine by cosine. It will be noted that the analysis of Gaumond and Jacobsen provides only the time dependence of both the backward and forward propagating strain waves, since the boundary conditions in the time domain are well defined. In an experiment in which the input ultrasonic wave is a pulse of duration T_1, their solutions give the peak echo amplitude when $T_2 >> T$ and when the interaction length over which the electric field is uniform is greater than (T_1+2T) times the acoustic velocity v. T_2 is the two-pulse decay time.

To summarize, the Mathieu equation approach to the general theory of parametric backward-wave echo generation predicts exponential growth of an ω-2ω echo, while the ω-ω echo may contain both unstable and stable contributions. When the pump is of low power and short duration, both hyperbolic sine and sine functions reduce to the value of their argument, giving the results quoted earlier that $u_B\propto E_0$ for an ω-2ω echo, and E_0^2 for ω-ω generation.

Damping has not been included in Eq. (2), but it is a straightforward matter to show that for a phenomenological attenuation coefficient α, the condition for exponential growth of ω-2ω echoes is

$$\tfrac{1}{4}C_1/C_0E_0\omega > \alpha v \ .$$

Most experiments to date have been carried out in the weak-coupling limit, for which the arguments of the functions in Eqs. (14a)–(14c) are much less than 1. For $LiNbO_3$, which is known to have large piezoelectric and electrostrictive coefficients, the ratios (C_1/C_0) and $(C_2/2C_0)$ are estimated to be 2.5×10^{-10} mV^{-1} and 10^{-19} m^2V^{-2}, respectively. For a pump frequency of 10 GHz at a peak power of 10^6 Vm^{-1} for 0.5 μs the arguments of the three functions given in Eq. (14) are estimated to be 2, 3×10^{-4}, and 1×10^{-3}, respectively. Thus deviations from the small coupling limit would be expected only in ω-2ω experiments carried out at high microwave frequencies and powers. We have therefore investigated this regime by us-

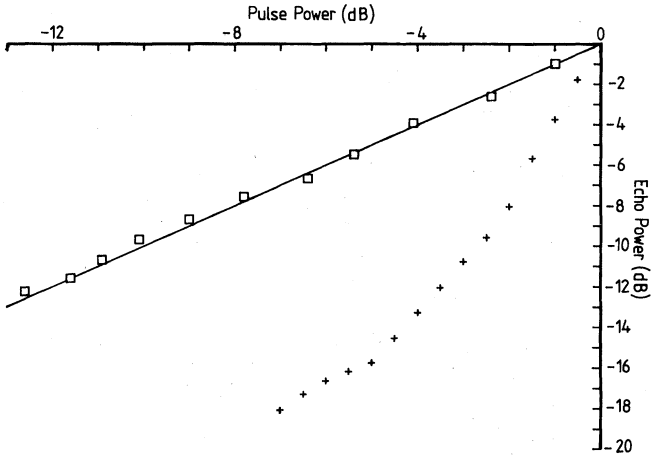

FIG. 2. Power dependences of the ω-2ω echo in X-cut $LiNbO_3$ expressed in dB. The squares are data for P_1 and the crosses for P_2. The straight line has a slope of unity.

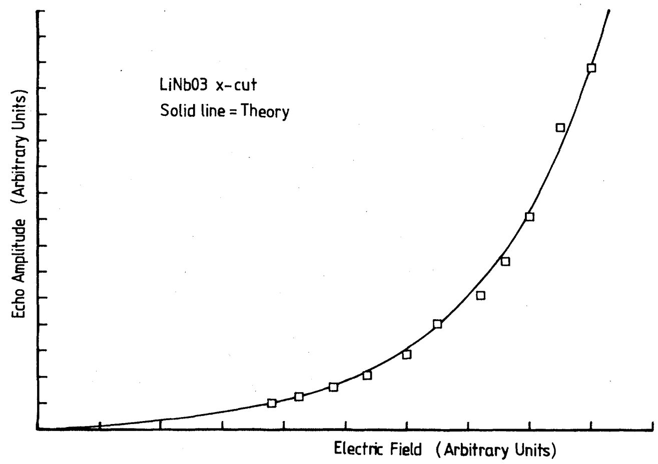

FIG. 3. Dependence of the echo amplitude expressed directly in terms of the pump electric field. The solid line is a best fit to a sinh x function.

ing a high-power pump operating at 35 GHz.

Full details of the experimental arrangements will be described elsewhere. The initial ultrasonic pulse was excited piezoelectrically at one end of the sample, a 2-mm-diam rod of either X or Z cut $LiNbO_3$ in a coaxial reentrant cavity which resonated at a frequency of 17.3 GHz. The remote end of the rod projected into the region of high electric field in a rectangular cavity, resonating at 34.6 GHz. By varying the length of the latter cavity, the electric field could be applied either parallel to or perpendicular to the rod axis. Pulse lengths were, respectively, 300 and 150 ns, with the separation time determined by the transit time for the primary ultrasonic pulse along the length of the sample. The maximum available powers were 1 and 5 kW, respectively. The latter figure for the electric field pump was limited only by electrical breakdown in the helium filled cavity at 1.3 K.

The significant data are the dependences of the backward-wave echo with respect to the amplitude (or power) of the two exciting pulses. In Fig. 2, we show the variation of the echo power, expressed on a logarithmic scale, with both the acoustic power P_1 which is proportional to the input microwave power, and the pump power P_2. Within the uncertainty of the power measurement, the P_1 variation has a slope of 1.03 ± 0.05, but the P_2 dependence of the echo only tends to unity at low-power levels and is strongly divergent at the highest-power levels. Similar data were obtained for both X and Z cut samples of $LiNbO_3$. When the experimental results are replotted on linear scales (Fig. 3), the variation of the echo amplitude agrees well with the $\sinh\theta$, where θ is proportional to the electric field E_0. The function was fitted to the experimental data at one arbitrary point and the best fit achieved by adjusting the constant of proportionality between θ and E_0. From a knowledge of the magnitude of the electric field in the cavity at the peak power and the intrinsic attenuation α, this procedure provides an estimate of the strength of the nonlinear coupling which is responsible for the echo generation. We obtain a value of $C_2 = 20 \pm 5\ \mathrm{C\,m^{-2}}$. This can be compared with the value obtained by Graham[12] for the coefficient g in Eq. (5) of $18 \pm 6\ \mathrm{C\,m^{-2}}$.

Parametric coupling between forward and backward ultrasonic waves in $LiNbO_3$ has also been observed by Thomson and Quate with a pump frequency of 2.8 GHz, but with much longer pulse lengths than those used in our experiments.[13] Their lower level of pump power was just sufficient for them to be able to detect the amplification of the forward wave and the onset of the exponential growth of backward-wave echo. In our experiments at the much higher microwave frequency, samples are insufficiently flat or parallel to allow measurements to be made on the forward waves.

In ω-ω experiments on both $LiNbO_3$ and $LiTaO_3$ with the signal, pump, and echo all at the same frequency of 17.3 GHz, deviations from linear power dependences were also observed at high-power levels, and there was some indication that the echo amplitude passed through a maximum as the pump power is increased. It was tempting to try to fit the data to the $\sin\theta$ function in (14b) which should describe this situation. However, the echo amplitude was not linearly dependent on the input signal in this region and a theory based on the simple-crystal Hamiltonian (1) cannot account for the magnitude and variety of effects that are observed. The origin of such echoes may well be due to impurities which are perturbed by a combination of electric and strain fields (Shiren *et al.*[3]).

In summary we have found that a Mathieu's equation approach to the parametric generation of phonon echoes in insulators predicts correctly the exponential growth observed at power levels beyond the small signal regime. We hope to explore also some other details of the Gaumond-Jacobsen model. Of particular interest will be the study of echo generation and signal amplification for experimental conditions which allow movement across the boundary between stable and unstable regions.

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