Phonon echoes and amplification of ultrasonic waves in a microwave electric field

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(Received 9 November 1976)

This paper presents a theoretical discussion of the phonon (two-pulse) echo and amplification of ultrasonic waves in a microwave electric field. A formula for the power of the phonon echo of Nth order is presented together with a formula giving the amplification of ultrasound in a microwave electric field. The relationship between the polarization of the phonon echo wave, the incident wave, and the direction of the externally applied electric field is pointed out. New predictions on the generation of phonon echoes whose frequency and polarization differ from the frequency and polarization of the primary wave are made.

I. INTRODUCTION

The phenomenon of polarization echo has been known since 1970 to have a macroscopic analogy to spin-echo effects in ferroelectric crystals.[1,2] Experimental investigation of this phenomenon has shown that there are two kinds of polarization echoes.[3-6]

The first kind of polarization echo is the so-called two-pulse echo.[3,4] The two-pulse echo of angular frequency ω is radiated from a piezoelectric sample at time $t=2\tau$ as a consequence of the fact that the sample has been excited by an rf pulse of the angular frequency ω at time $t=0$ and then by an rf pulse of angular frequency $2\omega/N$, where N is a whole number, at time $t=\tau$. The number N determines the order of the echo. Most experiments report echoes with $N=1$ or 2. However, two-pulse echoes of the higher orders have been also reported.[7] The generation of the echo from an ultrasonic wave in a microwave electric field is observed simultaneously with an amplification of the ultrasonic wave in the forward direction.[8]

The second kind of polarization echo is the three-pulse echo.[9,10] This echo is formed by three rf pulses of angular frequency ω applied to an investigated sample at time $t=0$, τ, and τ', and it is radiated at time $\tau+\tau'$. The three-pulse echoes in some cases exhibit an anomalously long relaxation time. The long relaxation time cannot be associated with dynamical properties of the medium and it has been proposed that the echoes of this kind are generated as a consequence of a static or stored polarization property of the sample.[9,10]

Thus only the two-pulse echo can be described as an echo of phonons. Theoretical discussions of the phonon echoes have used coupled-mode equations and have been limited to echoes of the first[3-11] and second order.[12] Moreover, the theory based on coupled-wave equations uses either a one-dimensional or a two-dimensional model. Therefore rather drastic simplifying assumptions occur for the interaction volume and the symmetry of the investigated crystal. Thus the present theory of the two-pulse echo is incomplete and should be, as it has been already suggested,[12] extended.

The problem of the phonon echo and amplification of ultrasonic waves in a microwave electric field will be studied in the present paper. We shall use the concept of phonon-photon interactions and time-dependent perturbation theory. Using the concept of phonons and photons the problem of phonon echo and amplification of ultrasound in a microwave electric field can be solved generally as a three-dimensional interaction.[13,14] This report gives formulas for the power of the Nth-order phonon echo and the amplification of an ultrasonic wave in a microwave electric field. The relationship between the polarization of the phonon-echo wave, the incident wave, and the direction of the applied field will be pointed out and the generation of the phonon echo which differs in frequency and polarization from that of the primary wave will be predicted.

II. PHONON ECHO

The generation of backward traveling phonons from an ultrasonic wave in a microwave electric field can be described as an N-photon two-phonon process using the following considerations.

If there occur photons of angular frequency ω_γ and wave vector $\vec{\gamma}$ in the volume of the investigated sample along with phonons of angular frequency ω_q and wave vector $\vec{q}$, then there is certain probability that N photons $(\vec{\gamma}, \omega_\gamma)$ annihilate while phonons $(\vec{q}, \omega_q)$ and $(\vec{q}^*, \omega_{q^*})$ are created. The law of conservation of energy and quasimomentum for the interacting quasiparticles requires

$$N\omega_\gamma = \omega_q + \omega_{q^*}, \quad N\vec{\gamma} = \vec{q} + \vec{q}^* . \tag{1}$$

Since ω_γ is in the region of ultrasonic frequencies, $N\vec{\gamma}$ is small compared to $\vec{q}$ and $\vec{q}^*$. Thus using (1) we can put $\vec{q}^* = -\vec{q}$. This means that the phonons

$(\vec{q}^*, \omega_{q^*})$ created through the nonlinear interaction of ultrasonic wave (phonons $\vec{q}, \omega_q$) with microwave electric field (photons $\vec{\gamma}, \omega_\gamma$) give rise to a backward traveling wave (phonon echo) while phonons $(\vec{q}, \omega_q)$ which have been created simultaneously with the echo phonons contribute to the forward traveling wave, which appears as an amplification.

A detailed calculation of the probability of the above-mentioned process requires the explicit form of the density of the interaction Hamiltonian operator for an N-photon two-phonon interaction. This can be obtained replacing the classical quantities by corresponding operators in a classical form of the interaction energy density of scattering of an ultrasonic wave in a microwave electric field. The classical form of the interaction energy density can be obtained from an expansion of the free-energy density of the investigated medium.[15] This, however, is not the easiest way because the externally applied electric field must be considered simultaneously with the field produced by the ultrasonic wave.[16]

Deriving the classical form of the interaction energy density for the scattering of the ultrasonic wave in the microwave electric field we will use the following straightforward consideration:

The ultrasonic wave propagating in a crystal has an energy density

$$U = \tfrac{1}{2} C_{ijkl} S_{ij} S_{kl}, \qquad (2)$$

where S_{ij} are the strain-tensor components and C_{ijkl} are elastic moduli "stiffened" by the electric field produced by the ultrasonic wave.[16] An externally applied electric field changes the energy density (1). This change can be phenomenologically described through the dependence of the moduli C_{ijkl} on the external electric field. Thus the ultrasonic wave acquires an energy density

$$U = \frac{1}{2}\left(C_{ijkl} + \frac{\partial C_{ijkl}}{\partial E_n} E_n + \frac{1}{2}\frac{\partial^2 C_{ijkl}}{\partial E_n \partial E_m} E_n E_m + \cdots + \frac{1}{N!}\frac{\partial^N C_{ijkl}}{\partial E_{n_1}\partial E_{n_2}\cdots\partial E_{n_N}} \times E_{n_1}E_{n_2}\cdots E_{n_N} + \cdots\right) S_{ij} S_{kl} \qquad (3)$$

when it propagates in the medium under the influence of an external electric field which has components E_n. Summation over repeated indices in relations (2) and (3) is understood.

Additional terms in relation (3) represent this part of the ultrasonic energy density which occurs as a consequence of the externally applied electric field. This is the interaction energy density. It can be shown that the Nth-order term in relation (3) gives in quantum mechanics the density of the interaction Hamiltonian for the N-photon two-phonon interactions. In deriving relation (3) the magnetoelastic coupling has been neglected. The nonlinear constants $\partial^N C/\partial E^N$ introduced in (3) reflect the dependence of the elastic properties on the external electric field. The constants $\partial^N C/\partial E^N$ are functions of C^E, e, and ϵ^s, where C^E stands for the elastic modulus at constant electric field, e is the piezoelectric stress constant, and ϵ^s is the dielectric constant at constant strain. These nonlinear constants for $N=1$ are the same as the coupling constants specified by Thompson and Quate.[8]

Using the strain tensor operator and the electric field operator[11] the interaction-energy-density operator-Hamiltonian for the N-photon two-phonon interactions is

$$\hat{H}_{\text{int}} = -\frac{i^N}{2\rho}\left(\frac{\hbar}{2V}\right)^{1+N/2} G_{q_1 q_2 \gamma_1 \gamma_2 \cdots \gamma_N}\left(\frac{\omega_{q_1}\omega_{q_2}\omega_{\gamma_1}\omega_{\gamma_2}\cdots\omega_{\gamma_N}}{v_{q_1}^2 v_{q_2}^2 \epsilon_{\gamma_1}\epsilon_{\gamma_2}\cdots\epsilon_{\gamma_N}}\right)^{1/2} (\hat{a}_{q_1} e^{i\vec{q}_1\cdot\vec{r}} - \hat{a}^*_{q_1} e^{-i\vec{q}_1\cdot\vec{r}}) (\hat{a}_{q_2} e^{i\vec{q}_2\cdot\vec{r}} - \hat{a}^*_{q_2} e^{-i\vec{q}_2\cdot\vec{r}}) \cdots (\hat{b}_{\gamma_N} e^{i\vec{\gamma}_N\cdot\vec{r}} - \hat{b}^*_{\gamma_N} e^{-i\vec{\gamma}_N\cdot\vec{r}}), \qquad (4)$$

where $\hbar$ is Planck's constant divided by 2π; $\hat{a}_q$, $\hat{a}^*_q$, $\hat{b}_\gamma$, and $\hat{b}^*_\gamma$ are annihilation and creation operators of phonons and photons, respectively, V is the volume of normalization (volume of the sample), ρ is the mass density, ω_q and ω_γ are angular frequencies of phonons and photons, v_q is the phase velocity of the ultrasonic wave $(\vec{q}, \omega_q)$, and ϵ_γ is

$$\epsilon_\gamma = k^{(\gamma)} \epsilon_{ij} k_j^{(\gamma)} \qquad (5)$$

while $G_{q_1 q_2 \gamma_1 \gamma_2 \cdots \gamma_N}$ is

$$G_{q_1 q_2 \cdots \gamma_N} = \frac{1}{N!}\frac{\partial^N C_{ijkl}}{\partial E_{n_1}\partial E_{n_2}\cdots\partial E_{n_N}} m_i^{(q_1)} m_k^{(q_2)} \times k_j^{(q_1)} k_l^{(q_2)} k_{n_1}^{(\gamma_1)} \cdots k_{n_N}^{(\gamma_N)}, \qquad (6)$$

where $m_i^{(q)}$ are components of the unit vector $\vec{q}/q$ and $k_i^{(q)}$ and $k_i^{(\gamma)}$ are components of polarization of the phonon $(\vec{q}, \omega_q)$ and photon $(\vec{\gamma}, \omega_\gamma)$, respectively. The indices γ and q in relations (4)–(6) are understood as double indices referring to both the polarization and the wave vector. Summations occur over the subscripted indices.

Let us assume that there are n_γ photons $(\vec{\gamma}, \omega_\gamma)$ and n_q phonons $(\vec{q}, \omega_q)$ in the volume of normalization and let us assume that the initial state of the phonons $(\vec{q}^*, \omega_{q^*})$ is empty. Thus the initial state is represented by the state vector $|n_q, 0, n_\gamma\rangle$. The total transition probability rate from this initial state to the final state $|n_q+1, 1, n_\gamma - N\rangle$ can be obtained in the form

$$\frac{dT}{dt} = \frac{\hbar^N G^2 L_x^2 L_y L_z}{2\rho^2 (2V)^{N+1}} \frac{\omega_q \omega_{q^*} \omega_\gamma^N}{v_q^2 v_{q^*}^3 \epsilon_\gamma^N} n_q n_\gamma^N , \tag{7}$$

where we have assumed that the interaction volume is a parallelepiped with edges L_x, L_y, and L_z, while the x axis is parallel to the wave vector $\vec{q}$. There is no summation over repeated indices q, q^*, and γ since they have been fixed with respect to the experimental situation.

The total transition probability rate (7) determines the average number of transitions per unit time. By multiplying (7) by $\hbar\omega_{q^*}$ the average power of the backward-traveling phonon echo wave is obtained in the form

$$P_{q^*} = \frac{G^2 L_x^2 \omega_{q^*}^2}{2^{N+2} \rho^2 v_q^3 v_{q^*}^3} P_q E_{0\gamma}^{2N} , \tag{8}$$

where the symbol P stands for the power of the ultransonic wave and $E_{0\gamma}$ is the amplitude of the external electric field.

III. PARAMETRIC AMPLIFICATION OF ULTRASONIC WAVES IN A MICROWAVE ELECTRIC FIELD

The phonon echo, as we have shown in Sec. II, is generated as a consequence of the annihilation of the photons $(\vec{\gamma}, \omega_\gamma)$ and creation of phonons $(\vec{q}, \omega_q)$ and $(\vec{q}^*, \omega_{q^*})$.

While phonons $(\vec{q}^*, \omega_{q^*})$ create a backward-traveling wave the phonons $(\vec{q}, \omega_q)$ contribute to the forward-traveling primary wave which appears as an amplification.[8]

In order to get the formula for the amplification of the forward-traveling wave $(\vec{q}, \omega_q)$ we shall use relation (7) which gives the average number of transitions per unit time and so determines the quantity dn_q/dt, where n_q is number of phonons $(\vec{q}, \omega_q)$ in the sample. If we take into account also the fact that phonons $(\vec{q}, \omega_q)$ are scattered due to ultrasonic attenuation in the sample, we can, using (7), write

$$\frac{1}{n_q} \frac{dn_q}{dt} = \frac{L_x G^2 \omega_q \omega_{q^*}}{2^{2N+2} \rho^2 v_q^2 v_{q^*}^3} E_{0\gamma}^{2N} - 2\alpha , \tag{9}$$

where α is a coefficient expressing the exponential decrease of the ultrasonic-wave amplitude per unit time.

Using relation (9) the power of the forward-traveling wave can be calculated

$$P_q(t) = P_q(0) \exp\left(\frac{G^2 \omega_q \omega_{q^*} L_x}{2^{2N+2} \rho^2 v_q^2 v_{q^*}^3} E_{0\gamma}^{2N} - 2\alpha\right) t , \tag{10}$$

where t is the interaction time and L_x is the length of the interaction volume in the direction $\vec{q}$.

IV. CONCLUDING REMARKS

We wish to point out several significant features of phonon echoes and the amplification of ultrasonic waves in a microwave electric field.

(i) Analyzing relation (1) we find that the frequency of the phonon echo is $\omega_{q^*} = \omega_q v_{q^*}/v_q$ while the frequency of the exciting electric field is $\omega_\gamma = \omega_q(1 + v_{q^*}/v_q)/N$. Since v_{q^*} can have three different values, three different kinds of phonon echoes of the Nth order can be generated from one primary wave $(\vec{q}, \omega_q)$. Similarly, there are, for a fixed N, three different frequencies of the exciting electric field for which the ultrasonic wave is amplified. The phonon echo of Nth order which differs both in frequency and polarization from the primary wave is a new prediction of the theory. The "new" echo of different polarization and frequency will not be observable in rough-cut samples. In the rough-cut samples only the backward wave of same polarization will retrace the initial-forward-wave path so as to arrive in phase at all points on the sample boundary. In order that the echoes of different polarization can be observed the initial ultrasonic wave must be launched from a single flat surface.

(ii) Relation (6) gives the relationship between the polarization of the phonon echo wave, the primary wave and the direction of the applied electric field together with the nonlinear coupling constant. The coupling constant for the echoes of Nth order is a $(N+4)$th-rank tensor. Since odd-rank tensors have zero values in centrosymmetric point groups, the echoes of odd orders can be observed only in noncentrosymmetric point groups.

(iii) Relation (10) shows that the total amplification of an ultrasonic wave in a microwave electric field results from a mutual competition between the amplifying effect of the electric field and the attenuation. The amplifying effect is proportional to L_x which is the length of the interaction volume. Thus the amplification of an ultrasonic pulse can also occur in materials which have relatively high attenuation provided that the sample is sufficiently long. Moreover, by amplifying the ultrasonic wave in the electric field of angular frequency ω_γ so that the echo wave has polarization different from the polarization of the forward-traveling primary wave, the interfering effects of the backward-traveling wave can be suppressed.

Finally, we would like to discuss the limits of applicability of our theory. We have assumed that the initial state of the backward-traveling wave is empty. This assumption does not limit the application of our formulas. Actually, if the initial phonon states $(\vec{q}^*, \omega_{q^*})$ were not empty backscattering [i.e., creaction of photons $(\vec{\gamma}, \omega_\gamma)$ through the annihilation of phonons $(\vec{q}, \omega_q)$ and $(\vec{q}^*, \omega_{q^*})$] should be considered simultaneously with the process of creation of the phonon echo. Formulas (8) and (10) could also be derived in this case provided that $n_\gamma \sim n_q \gg n_{q^*}$. It can be shown that our formulas hold with an accuracy of 2% even in the case when the amplitude of the backward-traveling wave is 10% of the amplitude of the incident wave.

Employing time-dependent perturbation theory it has been assumed that the phonon-photon coupling is weak. This assumption also applies in classical theory. Analyzing the experiments dealing with the phonon echo effects[7,8] it can be shown that the density of the interaction energy of ultrasonic wave and microwave electric field is very small compared to the energy densities of the incident ultrasonic wave and electric field even in materials with exceptional piezoelectric properties. This supports the weak-coupling assumption. Nevertheless, the assumption that the coupling is weak limits the applicability of the theory and should be kept in mind when experimental data are analyzed.

The phonon echo and the amplification of ultrasonic waves in microwave electric fields have great prospects in technical applications and also they give an excellent possibility to study elastoelectric anharmonicity of materials. We have discussed the above-mentioned phenomena from a uniform point of view showing that the phonon echo and the amplification of ultrasonic waves in microwave electric fields are related effects. Comparing our results with the results obtained previously[8] for $N = 1$ a very good agreement can be seen. This is quite valuable also because the results obtained previously have been based on a classical treatment using coupled-mode equations.

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