Acoustoelectric Interactions in Piezoelectric Semiconductors*

R. MAURO† AND W. C. WANG
Electrical Engineering Department, Polytechnic Institute of Brooklyn, Brooklyn, New York 11201
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Piezoelectric semiconductors such as cadmium sulfide exhibit a strong coupling between conduction electrons that are present in the substance and acoustic waves that are propagated along certain directions in the material. This energy exchange mechanism is highly nonlinear, and thus the simultaneous introduction of several collinear acoustic waves into the substance generates new signals at the conbination (sum and difference) frequencies. A theoretical explanation of this interaction mechanism, based on consideration of the nonlinear cross term present in the current-density equation, has been developed, and the validity of this method of analysis has been tested and qualitatively confirmed through experimentation.

I. INTRODUCTION

A STUDY of the propagation of ultrasonic waves in a substance provides substantial information about the electrical, mechanical, and thermal properties of the material. In particular, the use of finite-amplitude acoustic waves facilitates the investigation of the various interaction mechanisms present. These nonlinear effects are quite small in ordinary solids and rather difficult to detect experimentally.

For the case of piezoelectric semiconductors, however, as first noted by Hutson,[1] acoustic waves propagating in such materials can produce strongly coupled electric fields and space-charge waves with the nonlinear electron–electric-field interaction resulting in the generation of new electrical and acoustical signals.[2–5] This fact, together with the use of direct ultrasonic amplification in these substances, permits a significant increase in the signal-to-noise ratio for the observation of these nonlinear acoustic effects, and greatly simplifies the investigation of multiple wave interactions in these materials.

In order to describe these phenomena quantitatively, we have extended White's analysis[6] to obtain general expressions for both the acoustical and electrical signals that are generated as a result of the nonlinear interaction of several collinear acoustic waves of different frequencies. Further, experimental results are presented to illustrate the salient points of the theoretical study.

We will demonstrate that the derived general three-wave-interaction expressions for the acoustical signals can be readily reduced to those of Kroger[2] and Tell[3] on second-harmonic generation, and that the expression for the electrical signal can be reduced to Wang's[7] result for the acoustoelectric voltage. In addition, for acoustical signal generation, the phase-matching condition $\mathbf{k}_1+\mathbf{k}_2=\mathbf{k}_3$, which must be strictly observed in the case of ordinary parametric interactions, is shown to be relaxed in the amplifying media because of the participation of the drifted electrons. Further, we will establish that the generated electrical signals have amplitudes that are inversely proportional to their respective frequencies. As a result, the magnitude of the difference-frequency signal will be shown experimentally to be very much greater than those of the second-harmonic or sum frequencies.

II. DERIVATION OF PARTICLE DISPLACEMENT

Consider that, when a composite acoustical signal of frequencies ω_1 and ω_2 propagates in a properly oriented piezoelectric semiconductor, an ac longitudinal electric field is produced and travels with the acoustic wave. As the acoustic wave propagates, it will be progressively distorted because of the nonlinear interactions between the free carriers and the wave via the produced traveling ac electric field. With the assumption that the magnitudes of the second-harmonic and sum and difference frequencies are very much smaller than those of the fundamental frequencies ω_1 and ω_2, we present a small signal analysis that gives an estimation of the magnitude of the generated nonlinear components.

Following White's notation and derivation,[6] the particle displacement u, electric field E, carrier density n, electric displacement D, current density J, sound velocity v, wave vector k, and attenuation coefficient b are given as

$$u(x,t)=\sum_i A_i(x)e^{j(k_ix-\omega_it)}, \tag{1a}$$

$$E(x,t)=E_0+\sum_i E_i(x)e^{j(k_ix-\omega_it)}, \tag{1b}$$

$$n(x,t)=n_0+\sum_i n_i(x)e^{j(k_ix-\omega_it)}, \tag{1c}$$

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$$D(x,t)=D_0+\sum_i D_i(x)e^{j(k_ix-\omega_it)}, \tag{1d}$$

$$J(x,t)=J_0+\sum_i J_i(x,t)e^{j(k_ix-\omega_it)}, \tag{1e}$$

$$v_i=v_s\left[1+\frac{K^2}{2}\frac{1+\omega_c/\omega_D\gamma^2+\omega_i^2/\omega_D^2\gamma^2}{1+(\omega_c^2/\gamma^2\omega_i^2)(1+\omega_i^2/\omega_c\omega_D)^2}\right], \tag{1f}$$

$$k_i=\omega_i/v_i=k_{i0}-\Delta k_i, \tag{1g}$$

$$b_i=\tfrac{1}{2}K^2\left(\frac{\omega_c}{\gamma v_s}\right)\left[1+\left(\frac{\omega_c}{\gamma\omega_i}\right)^2\left(1+\frac{\omega_i^2}{\omega_c\omega_D}\right)^2\right]^{-1}, \tag{1h}$$

and

$$E_1=-\frac{e}{\epsilon}\frac{(\gamma+j\omega_1/\omega_D)S_1}{\gamma+j(\omega_c/\omega_1+\omega_1/\omega_D)}, \tag{1i}$$

where the conductivity-relaxation frequency $\omega_c=\sigma/\epsilon$, $(=2\pi F_c)$, the diffusion frequency $\omega_D=v_s^2/D_n$, the square of the electromechanical coupling coefficient $K^2=e^2/\epsilon c$, the nondispersive wave vector $k_{i0}=\omega_i/v_s$, and $\gamma=1-\mu E_0/v_s$, a measure of the relative electron drift velocity as compared to that of the acoustic wave.

Initially, let us consider the sum-frequency case $\omega_3=\omega_1+\omega_2$ and employ differentiation approximations similar to those used by Armstrong *et al.*,[8] who essentially assume that the loss or gain per acoustic wavelength is small. From the continuity equation, Gauss's law, and the definitions of current density, we may write

$$J_3=j\omega_3D_3=\mu q(n_0E_3+n_3E_0)+\mu q(n_1E_2+n_2E_1)+qD_n\frac{\partial n_3}{\partial x}. \tag{2}$$

Combining this result with the second piezoelectric equation of state $D=\epsilon E+eS$, we may express the electric field E_3 at the sum frequency in terms of the strain S_3 and the fundamental acoustic-signal-source term $n_1E_2+n_2E_1$ as

$$E_3=-\frac{e}{\epsilon}\frac{(\gamma+j\omega_3/\omega_D)S_3}{\gamma+j(\omega_c/\omega_3+\omega_3/\omega_D)}+\frac{\mu q(n_1E_2+n_2E_1)}{j\omega_3\epsilon[\gamma+j(\omega_c/\omega_3+\omega_3/\omega_D)]}. \tag{3}$$

Substituting Eqs. (1b) and (1c) into Eq. (3), and Eq. (3) into the first piezoelectric equation of state $T=cS-eE$, the modified wave equation is

$$\rho\frac{\partial^2u_3}{\partial t^2}-c_3\frac{\partial^2u_3}{\partial x^2}=\beta_3\left[\frac{\partial u_1}{\partial x}\frac{\partial^2u_2}{\partial x^2}+\frac{\partial u_2}{\partial x}\frac{\partial^2u_1}{\partial x^2}\right], \tag{4}$$

where c_3 is the effective second-order elastic constant,

$$c_3=c\left\{1+\frac{e^2}{\epsilon c}\left[\frac{\gamma+j\omega_3/\omega_D}{\gamma+j(\omega_c/\omega_3+\omega_3/\omega_D)}\right]\right\}, \tag{5}$$

and β_3 is the effective third-order elastic constant,

$$\beta_3=-j\frac{\mu e^3}{\epsilon^2v_s}\frac{\omega_c}{\omega_3}\frac{[\gamma+j\omega_1/\omega_D]+[\gamma+j\omega_2/\omega_D]}{[\gamma+j(\omega_c/\omega_1+\omega_1/\omega_D)][\gamma+j(\omega_c/\omega_2+\omega_2/\omega_D)][\gamma+j(\omega_c/\omega_3+\omega_3/\omega_D)]}. \tag{6}$$

It should be noted that, for $\gamma=0$, i.e., for drift velocity equal to the sound velocity, if we let $\omega_1=\omega_2$ and $\omega_3=\omega_{2H}=2\omega_1$, where ω_{2H} is the frequency of the second-harmonic signal, these results reduce to those obtained by Tell for the case of second-harmonic generation.

In order to solve Eq. (4), we assume that, for small incident strains, to a good approximation, the fundamental acoustic waves are unaffected by the nonlinearity. Thus, they may be represented by

$$u_1(x,t)=A_1e^{-b_1x}e^{j(k_1x-\omega_1t)}, \tag{7a}$$

$$u_2(x,t)=A_2e^{-b_2x}e^{j(k_2x-\omega_2t)}, \tag{7b}$$

where A_1 and A_2 are constants. In the remainder of the paper the imaginary part of Eq. (7) will be employed. Substituting Eq. (7) into (4), and employing the differentiation approximations,[8] the second-order differential equation may be reduced to the first-order differential equation

$$\frac{\partial u_3(x)}{\partial x}+b_3u_3(x)=\frac{\beta_3k_1k_2A_1A_2}{4c}\frac{k_1+k_2}{k_3}e^{-(b_1+b_2)x}e^{j(k_1+k_2-k_3)x}, \tag{8}$$

which, subject to the boundary condition $u_3(0,t)=0$, has the solution

$$u_3(x,t)=\left[\frac{\beta_3k_1k_2A_1A_2(k_1+k_2)\{e^{-b_3x}e^{jk_3x-\omega_3t}-e^{-(b_1+b_2)x}e^{j[(k_1+k_2)x-\omega_3t]}\}}{4ck_3\{[b_3-(b_1+b_2)]-j[k_3-(k_1+k_2)]\}}\right] \tag{9a}$$

$$=[A_3(x)e^{j(k_3x-\omega_3t)}], \tag{9b}$$

[8] J. A. Armstrong, N. Bloembergen, J. Dreuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).

or, taking the imaginary part of Eq. (9a), we obtain the actual solution

$$u_3(x,t)=\frac{|\beta_3|k_1k_2A_1A_2(k_1+k_2)\{e^{-b_3x}\cos(k_3x-\omega_3t+\Phi)-e^{-(b_1+b_2)x}\cos[(k_1+k_2)x-\omega_3t+\Phi]\}}{4ck_3[(b_3-b_2-b_1)^2+(k_3-k_2-k_1)^2]^{1/2}}, \tag{9c}$$

where Φ is the phase angle associated with $(-j)\beta_3$ and the complex denominator term in Eq. (9a). Note that in deriving Eq. (8), a correction factor of $-\frac{1}{2}j$ was included to account for the multiplication of the complex exponential terms. Kroger[2] has discussed the case of second-harmonic generation. His results are obtained by setting $\omega_1=\omega_2$, $k_1=k_2$, $b_1=b_2$, and $\omega_3=\omega_{2H}=2\omega_1$ in Eq. (9). To obtain the solutions for the difference-frequency case, merely replace ω_2 by $-\omega_2$ and k_2 by $-k_2$ in Eq. (9).

These solutions for the acoustic waves are quite similar to those obtained when considering finite-amplitude acoustic-wave propagation in lossy-dispersive nonpiezoelectric materials. However, in Eq. (9c), using, for example, the second-harmonic case for $\gamma<0$ (drift velocity greater than the sound velocity), the attenuation coefficients b will be negative, giving rise to an amplification of the second-harmonic acoustic wave. In general, $|b_{2H}|\neq|2b_1|$, so that one part of the solution will overtake the other. If γ is adjusted for the gain condition, and ω_c chosen so that $|b_{2H}|>|2b_1|$, the second harmonic produced initially near $x=0$ will be amplified at a rate greater than that at which new harmonic is being generated by the growing fundamental signal. For the opposite case, $|b_{2H}|<|2b_1|$, the second harmonic that is produced is locked in phase with the fundamental, and will travel and grow with it. Note in particular that, under gain conditions, beating (see the $\gamma<0$ curve of Fig. 1) does not occur in either case, since one of the sinusoidal terms in Eqs. (9) always grows out of proportion to the other. Thus, the rigidity of the selection rule $k_{2H}=2k_1$, so important for large harmonic production in lossless materials, is relaxed in this collinear amplifying case. For the sum- and difference-frequency cases, a similar, though not entirely analogous, argument may be employed. The variation of second-harmonic amplitude with distance is indicated in Fig. 1. In Fig. 2 we present the results of a typical interaction experiment; in particular, it illustrates the variation of the second-harmonic output with F_c and drift voltage γ, and agrees qualitatively with the theory developed in this section.

III. ELECTRICAL SIGNALS

In Sec. II, the nonlinear wave equation was solved for the particle displacement at the sum frequency to illustrate the form of the new acoustic waves that are generated. Now we will investigate the electrical signals of different frequencies that are generated across the piezoelectric semiconductor via the nonlinear interaction of the acoustic waves. By substituting Eq. (9b) into the wave equation

$$\rho\frac{\partial^2u_3}{\partial t^2}-c\frac{\partial^2u_3}{\partial x^2}=-e\frac{\partial E_3}{\partial x}, \tag{10}$$

and recalling that $A_3(x)$ varies very little per acoustic wavelength, we may readily show that

$$E_3(x,t)=(2c/e)(\partial A_3/\partial x)e^{j(k_{30}x-\omega_3t)}, \tag{11}$$

where $k_{30}=k_3+\Delta k_3$, as stated in Eq. (1g), and is the nondispersive wave vector at ω_3. The expression for the electric field is obtained by combining Eqs. (9a) and (11) to yield

$$E_3(x,t)=\frac{-j\beta_3k_1k_2(k_1+k_2)A_1A_2}{2ek_3[(b_3-b_2-b_1)+j(k_3-k_2-k_1)]}\times\{[(b_1+b_2)+j(\Delta k_1+\Delta k_2)]e^{-(b_1+b_2)x}e^{j[(k_1+k_2)x-\omega_3t]}-(b_3+j\Delta k_3)e^{-b_3x}e^{j[k_3x-\omega_3t]}\}. \tag{12}$$

Integrating the electric field along the length L of the crystal, the terminal voltage is obtained. Let us first examine a special case of the difference-frequency signal, that of the dc acoustoelectric voltage, for which ω_3, k_3, and b_3 are zero in Eq. (12), and ω_2, k_2, and Δk_2 in the equation are replaced by $-\omega_1$, $-k_1$, and $-\Delta k_1$, respectively. Equation (12) is then reduced to

$$E_{\mathrm{ac}}=-\frac{\mu e^2}{2e^2}\frac{[k_1A_1(0)]^2 2\gamma e^{-2b_1x}}{v_s[\gamma^2+(\omega_c/\omega_1+\omega_1/\omega_D)^2]}.$$

Integrating along the length of the crystal from 0 to L, and utilizing Eq. (1f) for b_1, one arrives at the result obtained by Wang, who used Weinreich's formulation.[9]

The general expressions for the terminal voltages at the sum and difference frequencies are lengthy. They are simplified, however, by multiplying by $1/jk_{i0}$ to approximate the integration of Eq. (12). This is valid

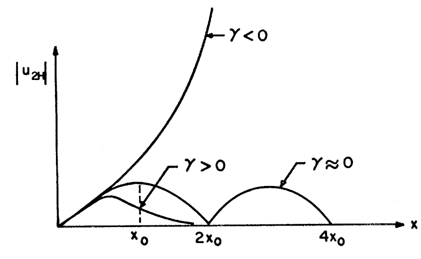

FIG. 1. Second-harmonic generation as a function of distance with drift voltage γ as a parameter ($x_0=\pi v_1^2/4\omega_1|v_1-v_{2H}|$).

[9] G. Weinreich, Phys. Rev. **107**, 317 (1957).

under the assumption that $k_{i0} \gg b_i$ and Δk_i, which is usually the case. If we further assume that the fundamental acoustic waves are completely attenuated at the $x=L$ boundary, and that no drift voltage is applied ($\gamma=1$), the voltage at the sum frequency ($\omega_3=\omega_1+\omega_2$), in particular, may be written

$$V_3=\left(\frac{R_L}{R_L+Z_{\text{in}}}\right)\left\{\frac{-jK^2\mu\omega_c}{\omega_3^2}\frac{(2+j\omega_3/\omega_D)[\frac{1}{2}cS_1(0)S_2(0)]e^{j\omega_3 t}}{[1+j(\omega_c/\omega_3+\omega_3/\omega_D)][1+j(\omega_c/\omega_1+\omega_1/\omega_D)][1+j(\omega_c/\omega_2+\omega_2/\omega_D)]}\right\}. \quad (13)$$

Note that the term $R_L/(R_L+Z_{\text{in}})$ represents the loading effect, where R_L is the external load resistor, and $Z_{\text{in}}=(G_0+j\omega C_0)^{-1}$ is the internal impedance of the crystal, with $G_0=\sigma_0 A/L$ and $C_0=A\epsilon/L$.

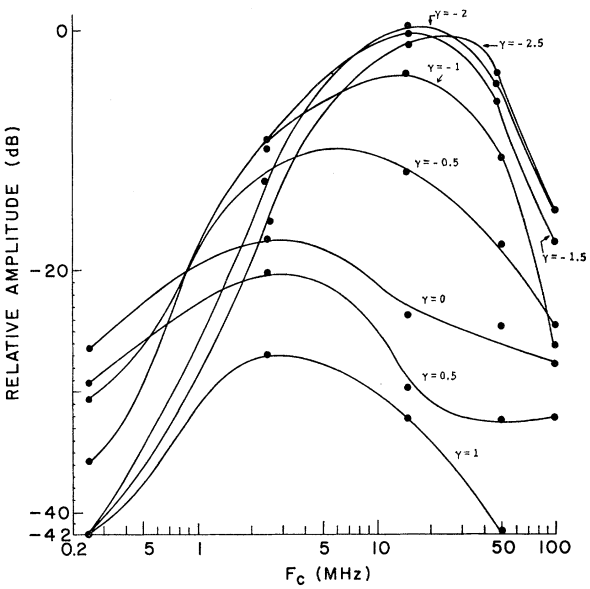

FIG. 2. Second-harmonic generation as a function of conductivity [$F_c=(1/2\pi)\sigma/\epsilon$], with drift voltage as a parameter.

Expressions similar to Eq. (13) can also be developed by direct analogy for the second-harmonic and difference-frequency amplitudes; these will be indicated by V_{2H} and V_d, respectively. In addition to the voltages at the sum and difference frequencies that are produced via the nonlinear interactions, there are terminal voltages induced by the fundamental waves at their respective frequencies ω_1 and ω_2. The magnitude of the voltage at ω_1, for example, is simply equal to the integral of Eq. (1i). Assuming complete attenuation of the fundamental wave at the $x=L$ boundary, we obtain

$$V_1=\left(\frac{R_L}{R_L+Z_{\text{in}}}\right)\left[\left(\frac{v_s}{\omega_1}\frac{eS_1(0)}{\epsilon}\right)\frac{1+j\omega_1/\omega_D}{1+j(\omega_c/\omega_1+\omega_1/\omega_D)}\right]e^{j\omega_1 t}.$$

Since the terminal voltages are approximately inversely proportional to their respective frequencies, the difference-frequency voltage V_d should be considerably larger than V_1, V_3, and V_{2H}. In order to compare the relative orders of magnitude of these signals to the dc acoustoelectric voltage V_{ac}, for simplicity neglect loading ($R_L\rightarrow\infty$) and diffusion, apply no drift voltage, and assume that the incident strain amplitudes at ω_1 and ω_2 are equal, i.e., $S_1(0)=S_2(0)$. If, for example, we further let $S_1(0)=10^{-5}$, and $\omega_2=\omega_1=20\omega_d$, then the relative signal magnitudes are given by $|V_d/V_{\text{ac}}|\approx 1$, $|V_3/V_{\text{ac}}|\cong 10^{-2}$, $|V_{2H}/V_{\text{ac}}|\cong 5\times 10^{-3}$, and $|V_1/V_{\text{ac}}|\cong 10^{-1}$.

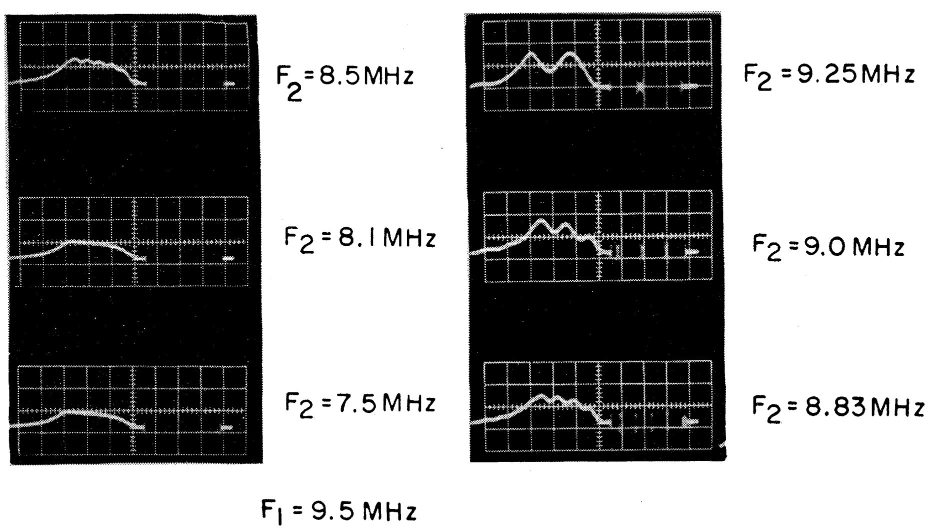

FIG. 3. Magnitude of the difference-frequency electrical signal as a function of the difference frequency (F_1-F_2). Arrows indicate the direction of increasing time (2 μsec per large scale division).

Thus, when the difference frequency ω_d is small compared to those of the fundamental signals, and the conductivity relaxation frequency ω_c is chosen to maximize the attenuation of the fundamental ($\omega_c \approx \omega_1$), the detected difference-frequency signal amplitude at ω_d will be comparable to that of the dc acoustoelectric voltage, and both will be much greater than the second-harmonic and sum-frequency amplitudes. The sequence of photographs presented in Fig. 3 illustrates the inverse frequency dependence of the difference frequency and, further, confirms that for small ω_d this detected voltage is comparable to its dc counterpart. This is to be expected since, in the limit as $\omega_d \rightarrow 0$, the difference-frequency acoustoelectric voltage becomes the dc acoustoelectric voltage.

These results describing the direct-voltage measurement of nonlinear acoustic-wave interactions in piezoelectric semiconductors may prove significant, since they indicate that an ultrasonic amplifier employed under gain conditions is capable of amplifying a modulated acoustic wave, detecting the modulation, filtering out the carrier and harmonics of the original signal, and converting the modulation portion of the acoustic wave directly into a large-amplitude voltage. This detected modulation signal is, of course, directly analogous to the aforementioned difference-frequency voltage.

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Carrier Lifetimes in Semiconductors with Two Interacting or Two Independent Recombination Levels

S. C. Choo
Westinghouse Research Laboratories, Pittsburgh, Pennsylvania 15235
(Received 16 July 1969)

Carrier recombination under (a) arbitrary steady-state and (b) small-signal near-equilibrium transient conditions has been studied theoretically for a two-interacting-level (ITL) model and a two-independent-level (IDL) model. Analytic solutions for carrier lifetimes have been obtained and manipulated into a form which facilitates comparison between the two models, as well as comparison between the steady-state and transient lifetimes as predicted by each model. It is shown that under small-signal steady-state and transient conditions the two interacting levels may be treated, with little loss of accuracy, as two independent levels, provided we describe the effective flaw density at each level by interacting-level equilibrium statistics. However, under appropriate conditions, the use of either ITL or IDL equilibrium statistics leads to essentially the same lifetimes; the ITL model is then indistinguishable from the IDL model. A comparison of the steady-state and transient lifetimes, whether of two interacting or two independent levels, shows that in certain circumstances the transient lifetime can exceed the sum of the steady-state electron and hole lifetimes, a possibility which does not exist if only one level is present. As a numerical example, the lifetimes in gold-doped silicon have been calculated and compared. Some possible applications of this work are proposed.

I. INTRODUCTION

The recombination statistics for holes and electrons through a set of single-level flaws have been treated extensively in the literature.[1–6] In the classic treatment by Shockley and Read,[1] steady-state solutions were obtained for the lifetime of electrons and holes. The extension to the small-signal[7] transient situation was given by Sandiford[3] and Wertheim[4] and recently by Sah[5] who, in applying the equivalent-circuit approach to single-level flaws, also examined the transient case of small signals superimposed on arbitrary steady-state conditions. As pointed out by Normura and Blakemore,[6] a complete analytic solution is not possible for transient decay involving signal levels and flaw densities of arbitrary magnitude; but some numerical calculations, with analytic approximations in various ranges, have been given by these authors.

An obvious extension to a set of single-level flaws is the case of two or more sets of single-level flaws acting in concert. Steady-state solutions for arbitrary flaw densities in a two-independent-level (IDL) model were obtained by Okada[8] and Kalashnikov,[9] while the small-signal transient solution was given by Wertheim[4] for n-type material with the restriction that the total density of flaws is less than that of the free carriers. The IDL case is reducible to the trapping model of Hornbeck

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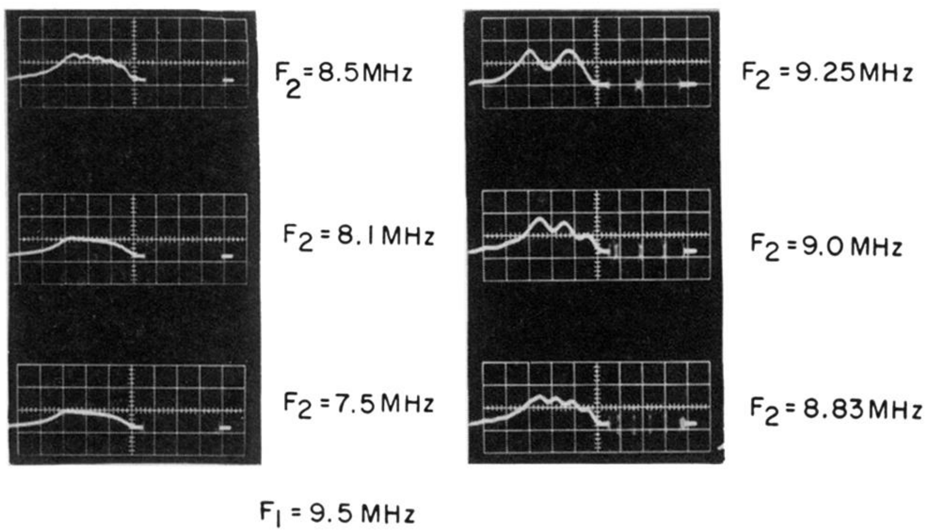

FIG. 3. Magnitude of the difference-frequency electrical signal as a function of the difference frequency (F_1-F_2). Arrows indicate the direction of increasing time (2 μsec per large scale division).