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AND RECOVERY OF X-CUT QUARTZ
UNDER SHOCK-WAVE COMPRESSION

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R. A. Graham
W. J. Halpin

SANDIA LABORATORIES



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Dielectric Breakdown and Recovery of X-Cut Quartz under Shock-Wave Compression*

R. A. GRAHAM AND W. J. HALPIN

Sandia Laboratory, Albuquerque, New Mexico 87115

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While a shock wave is traversing a disk of X-cut quartz, a piezoelectric current flows in an external circuit connected across the faces of the disk. In this paper measurements of this current are used to study dielectric breakdown and subsequent recovery which occurs in quartz. Quartz specimen disks were impacted at various stress levels in such a way as to produce shock waves that propagated along the X axis either in the direction of or opposite to that of the pressure-induced polarization. In the latter case, short-circuit current measurements show that breakdown occurs at a threshold stress greater than 10 and less than 13 kbar. Since the impact experiment produced one-dimensional electrical and mechanical conditions in the specimen disk, it was possible to formulate a mathematical model that permitted solutions for internal electrical fields and resistivity in terms of the measured current. Computations with this model show that the field in the stressed portion of the disk at breakdown is about 7.0×10^6 V/cm, which is an order of magnitude lower than the value observed at atmospheric pressure. Computations with the model also show that recovery from breakdown to essentially infinite values of resistivity occurs during the transit time of the shock wave when the field in the stressed region of the disk is "quenched" to a value of about 1.9×10^6 V/cm. This critical field appears to be the same for all shock stress levels investigated from 13 to 35 kbar. The dependence of the initiation of breakdown on the direction of wave propagation relative to the polarization direction indicates that the shock-wave front furnishes a source of free electrons.

INTRODUCTION

While a shock wave is traversing a piezoelectric disk, the stress-induced piezoelectric polarization produces electric fields which can approach the dielectric breakdown strength of the material at atmospheric pressure. Previously, we reported a detailed investigation of the piezoelectric, dielectric, and mechanical properties of X-cut quartz under shock-wave loading from 2 to 50

kbar.¹ We have also reported² that dielectric breakdown occurs if the quartz disk is oriented such that the polarization direction is opposite to the shock propagation direction (this is called the $-X$ orientation).³

¹ R. A. Graham, F. W. Neilson, and W. B. Benedick, *J. Appl. Phys.* **36**, 1775 (1965).

² R. A. Graham, *J. Appl. Phys.* **33**, 1755 (1962).

³ The $-X$ orientation is obtained when the X-cut crystal disk is oriented such that the shock wave propagates in a direction opposite to that of the electrical polarization produced, that is, from the $+X$ electrode to the $-X$ electrode. The polarity of the electrodes is determined by placing the disk under compression and noting the direction of current flow through a high-impedance load.

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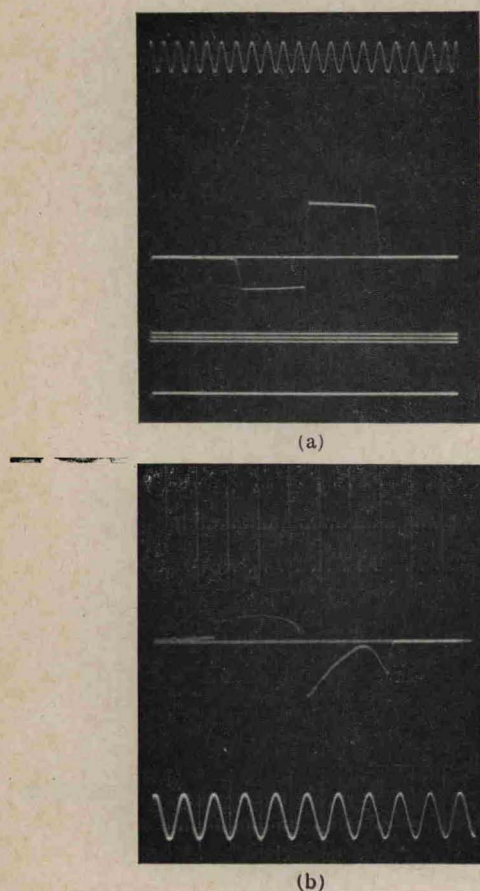


FIG. 1. Typical current-time records obtained from (a) $+X$ orientation disks and (b) $-X$ orientation. Time increases from right to left. The records show the current resulting from two wave transits through the disk; the first portion of the pulse corresponds to the propagation of the impact-induced shock wave, and the second portion corresponds to the reflected pulse from the rear of the disk. The analysis is concerned only with the first wave transit. The $+X$ orientation records show no evidence of breakdown while the $-X$ orientation records show a large reduction in current resulting from dielectric breakdown when the shock wave enters the disk. The upper sinusoidal traces in (a) and (b) are 10 MHz timing waves. The lower traces are for current calibration.

Because of the simultaneous combination of stress and high values of electric field that occurs under shock loading, the shock-wave experiment provides a novel opportunity for studying the effect of stress on the dielectric strength, a subject on which there is very little data in the literature.⁴

Measurements of current in an external short circuit connected across the faces of a shock-loaded quartz disk can be used to study this dielectric breakdown behavior. When breakdown occurs, this current is characterized by an initial jump as the shock wave enters the disk, followed by a sharp drop as the high field occurring at this time initiates the breakdown process. Later in time, when the field has been quenched, there is a tendency for the current to recover. Since the previous $-X$ orientation current-time measurements²

⁴ S. Whitehead, *Dielectric Strength of Solids* (Clarendon Press, Oxford, England, 1953).

were taken on a specimen whose geometry resulted in three-dimensional electrical and mechanical conditions, it was not possible to obtain a satisfactory mathematical description of the physical processes involved. However, we have now conducted experiments in a one-dimensional guard-ring geometry and observed the short-circuit current during breakdown. For these experiments a mathematical description has been possible, and with it we have performed a quantitative analysis of the experimental data to obtain the transient resistivity-time and resistivity-field behavior accompanying the dielectric breakdown and recovery. It is the object of this paper to report these results.

We will first describe the experiments and the data obtained from them. Following this, a mathematical model for analyzing variable resistivity will be developed. Finally, the model will be applied to the experimental data and the results discussed.

EXPERIMENTAL OBSERVATIONS

The experiment for measuring the short-circuit current from shock-loaded quartz disks has been described previously.¹ Briefly, a projectile faced with a quartz disk is propelled down the barrel of a compressed gas gun to impact on a target that contains the short-circuited specimen disk. The impact of the two disks causes a shock wave to move into the specimen, and the amplitude of the wave is determined by the known mechanical properties of quartz and the projectile velocity, which is accurately measured. The specimen disk has a guard-ring electrode configuration to insure one-dimensional conditions, and special precautions are taken to prevent breakdown along external surfaces. Details of the specimen configuration have been described previously.^{1,5} The current is obtained from the voltage drop across a known resistance which is connected across the electrode faces of the specimen.

The current-time record shown in Fig. 1(a) is characteristic of the output of disks having an orientation such that the shock wave propagates from the $-X$ polarity electrode to the $+X$ polarity electrode. No breakdown occurs in this orientation up to a shock stress of at least 30 kbar as evidenced by the approximately constant value of the current. As shown in Fig. 1(b), however, disks oriented such that the shock wave travels from the $+X$ electrode to the $-X$ electrode exhibit current-time records that deviate considerably from constant current.

Experimental current-time data were obtained in the $-X$ orientation for stresses ranging from 8 to 35 kbar. Experiments at 8 and 10 kbar showed no breakdown. The first indication of breakdown in the specimens occurred at 13 kbar. As the input stress was increased above 15 kbar, the breakdown became more pronounced with the current dropping to zero in times

⁵ W. J. Halpin, O. E. Jones, and R. A. Graham in *Symposium on Dynamic Behavior of Materials*, ASTM Special Technical Publication No. 336 (American Society for Testing Materials, Philadelphia, 1963).

of about 10^{-7} sec. After this time the current was observed to recover (increase in magnitude) to varying degrees depending on the value of the stress. For higher stress the current showed less recovery from the breakdown.

Luminescence observations were also made during breakdown, and these observations correlate with the current pulse results in a qualitative way.⁶ Previous luminescence results⁷ were not applicable to the present problem since they were obtained at stresses greater than those used in this investigation. These photographs⁸ (Fig. 2) showed that the breakdown occurs at discrete points in the crystal with a luminous region growing around the points. The last frame shows that the intensity of the luminescence decreases after about 200 nsec. The records demonstrate that there is good

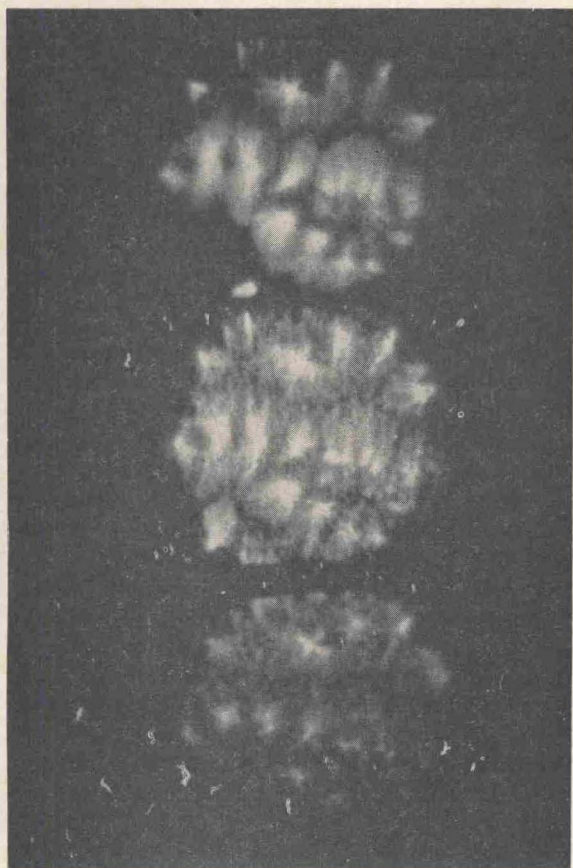


FIG. 2. Image converter framing camera record of the luminescence accompanying dielectric breakdown in $-X$ orientation disks at 19 kbar. The photographs are taken at three different times, looking down the X axis of the disk as the shock wave moves toward the observer. The first (upper) frame is taken within 50 nsec after impact and the following two frames are taken at 100 nsec intervals thereafter.

⁶ The luminescence observations are conducted on disks in an open-circuit condition. This does not affect the qualitative comparison between the current-time measurement and the luminescence observations since at the time the shock wave enters the disk the field in the stressed region is the same under both conditions.

⁷ W. P. Brooks, J. Appl. Phys. **36**, 2788 (1965).

⁸ This experiment was performed by G. E. Ingram with a Space Technology Laboratories image converter camera.

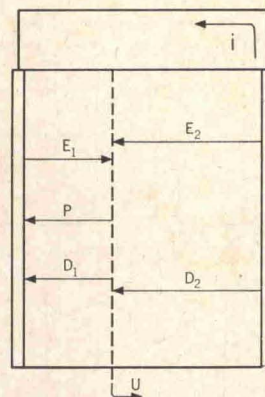


FIG. 3. Electric vectors produced in piezoelectric quartz under shock-wave loading. The position of the shock front is indicated by the dashed line. The vector arrows indicate those directions that are defined as positive in the analysis. The polarization direction shown is opposite to the propagation direction and depicts the $-X$ orientation.

qualitative correlation between the current-time and the luminosity-time behavior, as might be expected for a dielectric breakdown.

Attempts to analyze the complete current-time profile on the basis of a finite constant resistivity were unsuccessful and led to the development of a mathematical model which allows for a variable resistivity under shock-wave loading.⁹

MATHEMATICAL MODEL

In Fig. 3 we consider a shock wave having a plane front and a step profile in time and space as it traverses an X -cut quartz disk of thickness l . The faces of the disk each have area A and are covered with electrodes that are connected by a short-circuit. The wave propagates from left to right with a constant velocity U in the direction of the X axis and with its front perpendicular to that axis. The region of the disk that comes under stress experiences a piezoelectric polarization P , determined by the product of the stress and the appropriate piezoelectric coefficient (the direction shown for P in Fig. 3 is indicative of the $-X$ orientation for the disk). As the wave moves through the disk, the region of polarization continually lengthens, and this causes a current i to flow in the externally connected short-circuit. The process is accompanied by fields E_1 across the stressed region and E_2 across the unstressed region, which vary with time and are oppositely directed for voltage balance.

Infinite Resistivity

We now investigate the time variations of the current i and the electric fields E_1 and E_2 mathematically. In order to provide background for interpreting the current pulses, we will first digress and review the conditions existing for infinite resistivity where break-

⁹ Analysis of the effects of shock-wave-induced finite, constant resistivity has been reported by R. H. Wittekindt, Harry Diamond Laboratories Rept. TR 922, May 1961, and Y. B. Zel'dovich, Sov. Phys.—JETP **26**, 159 (1968).

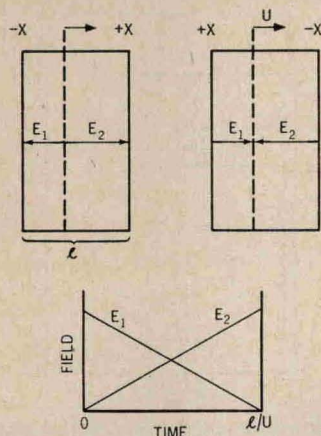


FIG. 4. Electric field diagrams for shock-loaded quartz. The upper diagrams show the directions of the electric fields E_1 and E_2 for the $+X$ (left) and $-X$ (right) orientations. The lower diagram shows the variation of E_1 and E_2 with time for the case of infinite resistivity and equal permittivities for the stressed and unstressed regions. The field E_1 in the stressed region is maximum when the shock wave enters the disk, while the field in the unstressed region E_2 is maximum at shock transit time $t=l/U$. The maximum fields are directly proportional to the magnitude of the shock stress and are equal to 5.3×10^6 V/cm at 10 kbar.

down does not occur. In Fig. 3 the shock-wave front is shown at a time t measured from the instant it entered the disk on the left, and the polarization, electric displacement, and electric field vectors are shown in directions that are defined as positive. These vectors are assumed to be uniform in magnitude over the face of the disk with directions parallel to its axis, and the subscripts 1 and 2 refer as above to the stressed and unstressed regions, respectively. Let ϵ_1 and ϵ_2 be permittivities for the two regions. We can then write the dielectric equations as

$$D_1 = -\epsilon_1 E_1 + P \quad (1)$$

and

$$D_2 = \epsilon_2 E_2. \quad (2)$$

The short-circuit condition allows us to employ Kirchoff's law and write

$$E_1 U t = E_2 (l - U t) \quad \text{for } 0 \leq t \leq l/U. \quad (3)$$

With the assumption of infinite resistivity, it follows that the current (positive direction shown in the figure) is given by

$$i = -A (dD_2/dt), \quad (4)$$

and also that $D_1 = D_2$. With the latter condition Eqs. (1)–(3) can be solved for D_2 , with the result that

$$D_2 = \epsilon_2 P U t / [\epsilon_2 U t + \epsilon_1 (l - U t)]. \quad (5)$$

We then differentiate (5) with respect to t and substitute in (4) to get

$$i = -\alpha A U P l / [U t + \alpha (l - U t)]^2, \quad 0 < t < l/U, \quad (6)$$

where $\alpha = \epsilon_1/\epsilon_2$. The analysis so far shows that if the resistivity is infinite and if $\alpha = 1$, a constant current will result from a step-function stress imparted to the crystal.¹⁰ The current-time waveforms observed in the $+X$ orientation and shown in Fig. 1(a) show a close correspondence to that predicted from Eq. (6).

From Eqs. (2), (3), and (5), the electric fields E_1 and E_2 resulting from the stress-induced polarization are found to be given by

$$E_1 = P(l - U t) / [\epsilon_2 U t + \epsilon_1 (l - U t)],$$

$$E_2 = P U t / [\epsilon_2 U t + \epsilon_1 (l - U t)]. \quad (7)$$

Thus, as shown in Fig. 4, the field in the stressed region of the disk is maximum at the time the shock wave enters the disk, and then decreases linearly (for $\alpha = 1$) in time to a value of zero at wave transit time, which is of the order of 1 μ sec. On the other hand, the field in the unstressed region is zero when the shock wave enters the crystal and increases to a maximum value at wave transit time. When the direction of propagation of the shock wave is reversed relative to the polarity of the X axis, the field directions are reversed.

The distinctly different field-time environment in the stressed and unstressed regions of the disk permits identification of the region through which the breakdown occurs. Breakdown is observed to occur early in time, which corresponds to the time of maximum field in the stressed region. Hence, breakdown occurs through the stressed region.

Finite Resistivity

We now attempt to obtain a quantitative interpretation of recovery from breakdown observed in the $-X$ orientation experiments by developing a mathematical model which includes the effect of a variable resistivity. First, a very general assumption is made that some process occurs uniformly throughout the stressed region 1 in such a way as to alter the rate at which D_1 changes as a function of time. We express this effect by introducing a loss factor F in the equation

$$dD_2/dt = (dD_1/dt) - F(t), \quad (8)$$

which now, in general, relates D_1 and D_2 in a way other

¹⁰ If $\alpha < 1$ the current value at $t=0$ is greater than the value at $t=l/U$ while for $\alpha > 1$ the opposite is true. However, as can be seen by solving Eq. (6) for

$$\int_0^{l/U} i(t) dt,$$

the charge Q that passes through the short circuit from $t=0$ to $t=l/U$ is equal to PA , independent of the value of α for this case of infinite resistivity. Thus, in an experimental result, the deviation of the charge Q from the value PA is a measure of the extent of the breakdown or of the average finite resistivity of the disk during wave transit time.

than $D_1 = D_2$. In the physical sense, a positive value for F at some time t would indicate a net *rate* of loss of volume polarization in region 1 (e.g., a conduction current) while a negative value would indicate a net *rate* of gain. We now make the substitutions from Eqs. (1)–(3) for D_1 in (8) and take a derivative to obtain the differential equation

$$-dD_2/dt = UtF/[Ut + \alpha(l - Ut)] \\ -\alpha l D_2/t[Ut + \alpha(l - Ut)]. \quad (9)$$

By assuming different functions F and certain initial conditions, it is possible to solve (9) for D_2 and then i in an attempt to fit the experimental results. However, since i is already known experimentally, a more direct approach is to obtain F in terms of i . We do this by solving (9) for F , and substituting for D_2 and dD_2/dt

$$\rho_1(t) = t(l - Ut) \int_0^t i(\bar{t}) d\bar{t} / \{ \epsilon_1 l \int_0^t i(\bar{t}) d\bar{t} - i l [\epsilon_2 Ut + \epsilon_1(l - Ut)] \}. \quad (12)$$

Similarly, we can solve for E_1 from Eqs. (2)–(4) to get

$$E_1(t) = -[(l - Ut)/Ut\epsilon_2 A] \int_0^t i(\bar{t}) d\bar{t}. \quad (13)$$

Using these equations, we can obtain numerical solu-

from (4) to get

$$F(t) = \left\{ \frac{[\alpha(l - Ut) + Ut]}{Ut} \right\} (i/A) \\ - (\alpha l / AU^2) \int_0^t i(\bar{t}) d\bar{t}. \quad (10)$$

There remains, however, the problem of interpreting the meaning of a function F in terms of known physical processes. We do this by relating F to the resistivity in the stressed region ρ_1 and field E_1 by the expression

$$F = E_1/\rho_1. \quad (11)$$

In this way F represents a conduction current due to a field E_1 impressed on a material of resistivity ρ_1 . From Eqs. (2)–(4), (10), and (11), we can solve for ρ_1 to get

tions for ρ_1 and E_1 as functions of time by substituting experimentally determined values of current taken from the observed current–time records. This enables the resistivity to be calculated for each experiment as the field is reduced and recovery from breakdown occurs.

RESULTS AND DISCUSSION

The first indication of breakdown was observed in the $-X$ orientation at a stress of 13 kbar. The maximum field at this stress as computed from Eq. (7) (for $t=0$) is 7.0×10^5 V/cm.¹¹ This field is almost an order of magnitude smaller than the field required for breakdown at atmospheric pressure.¹²

Resistivity ρ_1 vs time and field E_1 vs time, computed by applying Eqs. (12) and (13) to the current–time pulses obtained at stresses of 15 and 26 kbar, are plotted in Fig. 5. The more rapid reduction of E_1 with time shown in the 26 kbar record should be compared with the linear decrease in time to be expected when resistivity is infinite (and $\alpha=1$) (see Fig. 4). The behavior of ρ_1 is seen to be highly time dependent, thus demonstrating that analysis of the current–pulse data based on the assumption of constant resistivity would be of little value. The main features of the ρ_1 vs time curves are a rapid reduction in resistivity when breakdown occurs, followed by a recovery later in time when the field is “quenched” to some lower value.

Graphs of resistivity ρ_1 vs field E_1 are shown in Fig. 6 for various values of stress from 13 to 36 kbar. Although

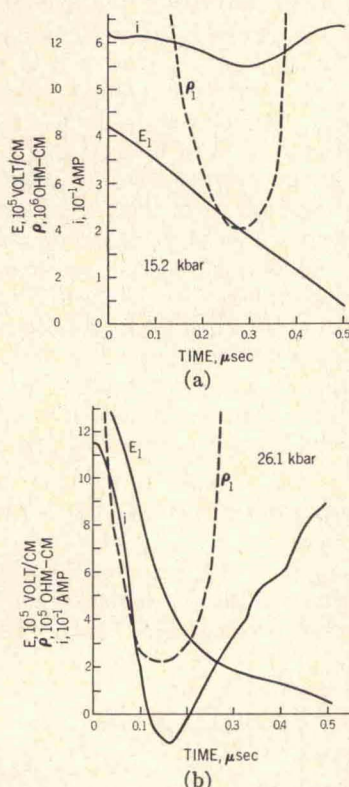


FIG. 5. Typical current–time records for the $-X$ orientation with the resulting calculated resistivity–time and field–time behavior. The current–time records are adjusted to compensate for the short but finite risetime of the current pulse resulting from impact “tilt.”

¹¹ For this computation, ϵ_1 is taken as 4.00×10^{-13} F/cm from the atmospheric pressure value of R. Bechmann, Phys. Rev. **110**, 1060 (1958), and the observed 0.3% increase in permittivity with stress reported in Ref. 1. The polarization, P , is also taken from Ref. 1 as 2.78×10^{-7} C/cm².

¹² A. Von Hippel and R. J. Maurer, Phys. Rev. **59**, 820 (1941).

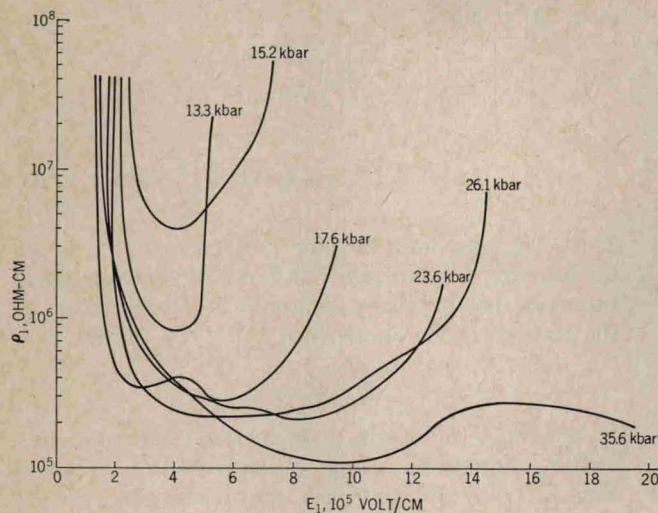


FIG. 6. Resistivity vs field calculated from the observed current-time records. Breakdown occurs at field values to the right of the figure while recovery occurs later in time as the field decreases to values on the left. At all stress levels the resistivity calculated from the observed current-time waveforms is seen to approach an infinite value ($>4 \times 10^7 \Omega \cdot \text{cm}$) when the field is reduced to $1.9 \pm 0.5 \times 10^5 \text{ V/cm}$.

each experiment shows behavior of a somewhat different character, there is one distinct feature of the recovery which is common to all experiments. Recovery of essentially infinite values of resistivity ($>4 \times 10^7 \Omega \cdot \text{cm}$) occurs over a surprisingly narrow range of fields for all stress levels. Thus, it appears that there is a stress-independent critical field of $1.9 \pm 0.5 \times 10^5 \text{ V/cm}$ for the recovery of quartz from dielectric breakdown.

The solutions for ρ_1 at later times after complete recovery give negative values (not shown in the figure) which indicate that a net gain in the rate of electric displacement occurs after recovery becomes well established. This represents the return of previously "shorted" material to a polarized state, which is the expected behavior.

These results describe two phases of the dielectric breakdown phenomena which are: (1) the initiation of the breakdown and (2) the recovery that follows when the electric field is quenched. In shock-loaded *X*-cut quartz, breakdown occurs in the $-X$ orientation, but when the field polarity is reversed, that is, when the quartz is in the $+X$ orientation, breakdown does not occur. Since the field that is present during breakdown is in a direction to accelerate electrons from the shock-wave front into the stressed region of the quartz, it appears that breakdown is initiated from a source of free electrons at the shock-wave front. The presence of free electrons at the front requires shock-induced electronic energy level changes which cannot be accounted for by the magnitude of the strain energy behind the shock wave, although they can easily be accounted for by the motion of dislocations in the wave front. Apparently, the high shear stress in the wave front causes localized dislocation motion, even though

the stress behind the wave is considerably less than the Hugoniot elastic limit^{13,14} of about 40 kbar.

The phenomenon of a dependence of conduction processes under shock loading on the direction of the stress-induced electric field is not restricted to quartz alone. Recently, for example, Cutchen¹⁵ has shown the effect to occur in ferroelectric ceramics.

The results of the analysis of the recovery process after breakdown in shock-loaded quartz show that the recovery of high-resistivity values occurs when the electric field is quenched to a value of about $1.9 \times 10^5 \text{ V/cm}$. Since this value of field is characteristic of all shock stress levels from 13 to 36 kbar, it appears there is a critical field for recovery which is independent of the stress. The experiments at the various stress levels produced different values of piezoelectric polarization in the quartz specimens so that upon breakdown different values of current were involved. Thus, for the range of currents in the experiment, the critical field for recovery also appears to be independent of the current. Because the critical field determined here does not depend on stress, it may be characteristic of recovery from dielectric breakdown at atmospheric pressure.

ACKNOWLEDGMENT

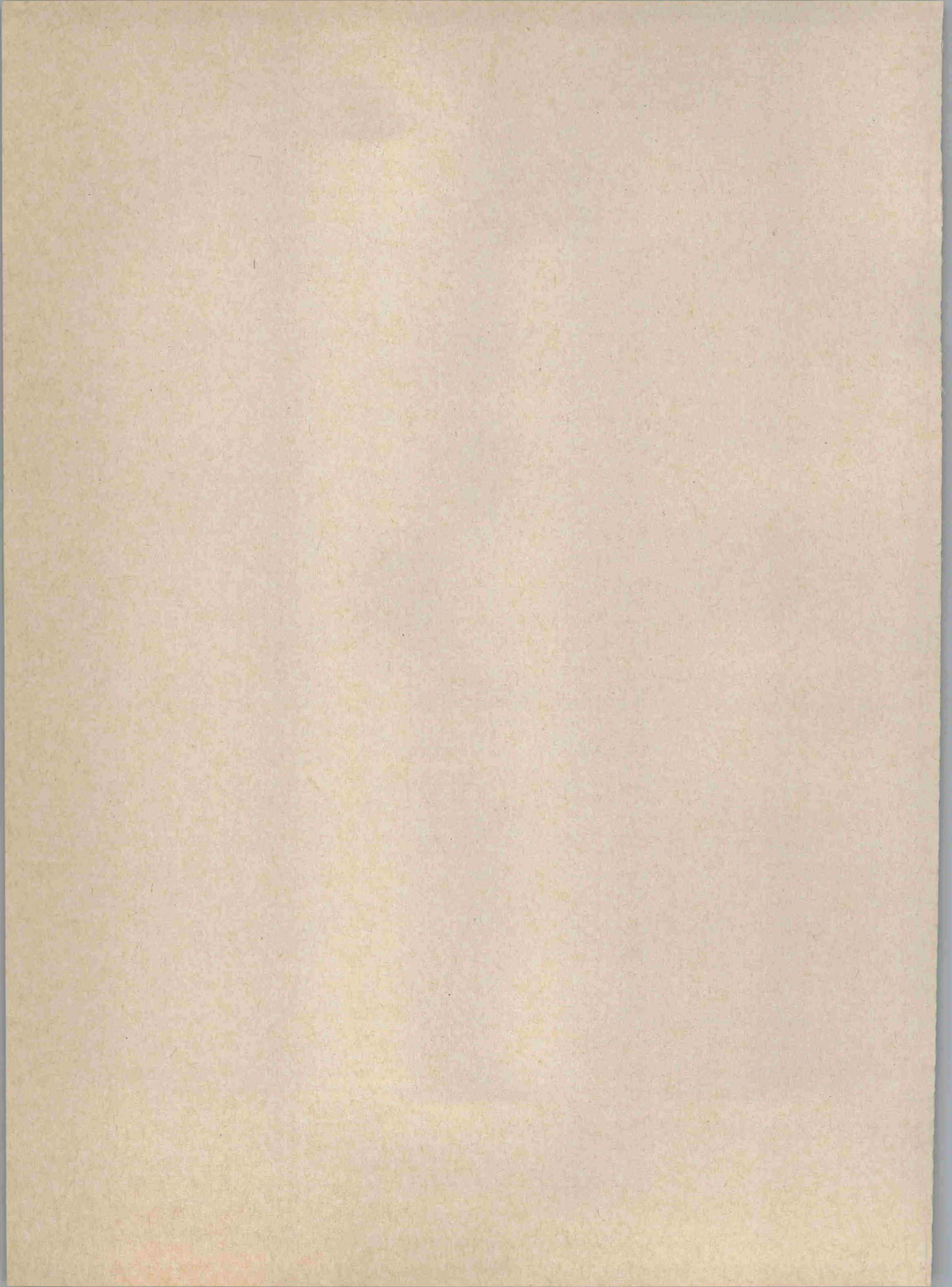
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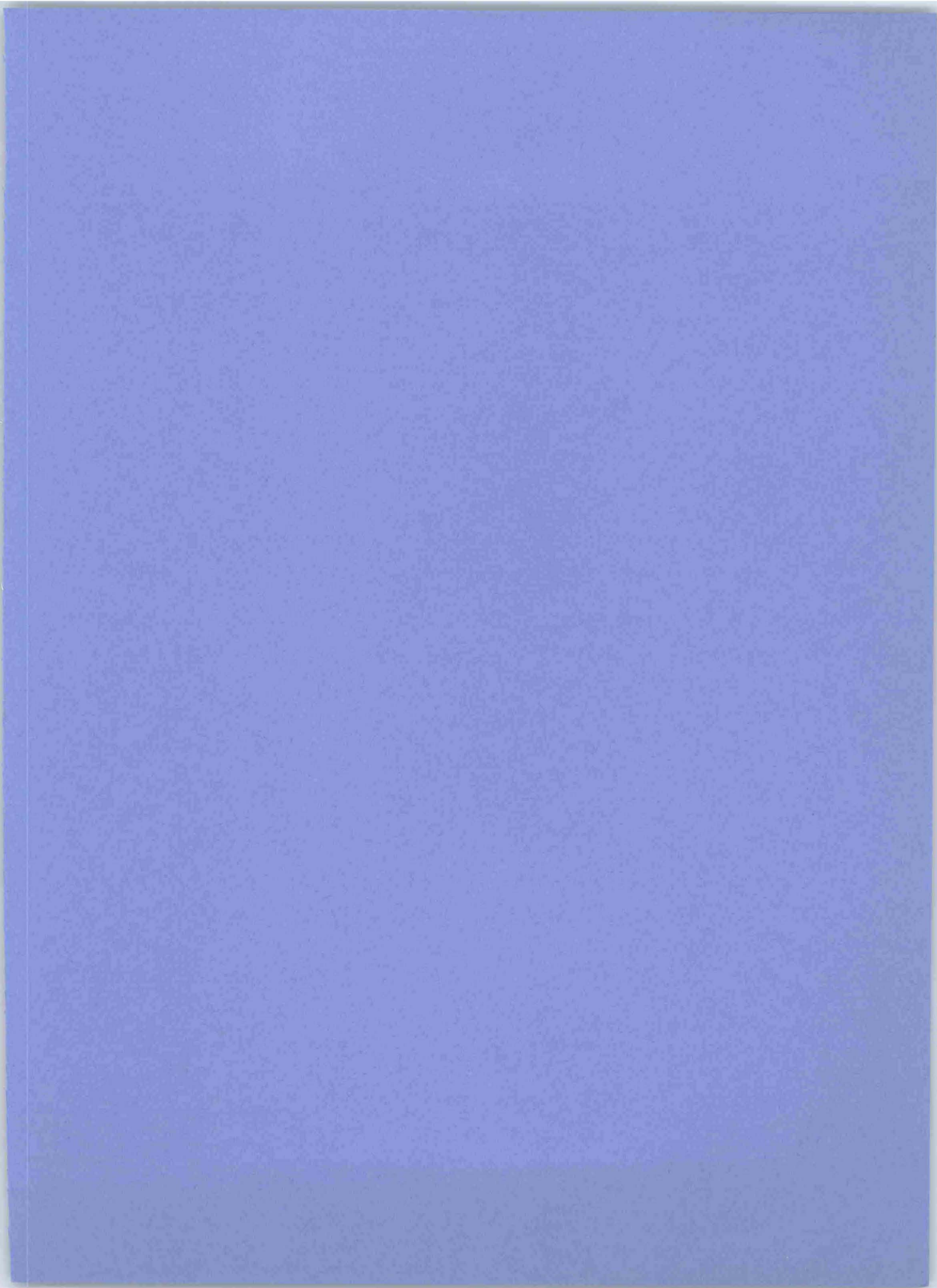
¹³ Jerry Wackerle, *J. Appl. Phys.* **33**, 922 (1962).

¹⁴ Richard Fowles, *J. Geophys. Res.* **72**, 5729 (1967).

¹⁵ J. Thomas Cutchen, *J. Appl. Phys.* **37**, 4745 (1966).

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