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by

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Dielectric Anomaly in Quartz for High Transient Stress and Field*

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Neilson and Benedick have reported an anomalous piezoelectric behavior for negatively-oriented synthetic alpha-quartz crystals when subjected to transient stress of about 65 kbar. This paper describes the piezoelectric behavior of negatively-oriented quartz in the stress region of from 5 to 50 kbar. The first indication of anomalous behavior occurs at 8 kbar. Between 8 and 24 kbar the negative x-current waveforms show evidence of partial electric breakdown in the quartz. Between 25 and 34 kbar disruptive breakdown occurs. Above 34 kbar disruptive breakdown is followed by gross conduction with positive currents being observed for stress greater than about 50 kbar. The fields associated with the piezoelectric behavior are lower than the field for steady-state electric breakdown at atmospheric pressure. It is proposed that the anomaly is triggered by stress-induced dislocation motion resulting in liberated electrons which are accelerated into the stressed region of the specimen by the high negative electric field.

INTRODUCTION

IN several previous papers[1,2] results of experiments were presented in which the piezoelectric behavior of synthetic alpha-quartz was observed under high transient stress. The technique[3] involves impacting flat-faced cylinders of quartz crystals upon each other with precise control being maintained on alignment. By using the Hugoniot conservation of momentum[4] and the mechanics relations of flat cylinder impact, the stress due to impact may be computed from the experimentally determined impact velocity. The experiments produce transient stress in the range of 5 kbar to about 50 kbar. The piezoelectric behavior of the impacted quartz specimen during the initial transient of the stress wave is observed as short-circuited current external to the crystal.

X-cut crystals may be oriented so that the stress wave produced by the impact travels from the −X face to the +X face or from the +X face to the −X face. The sign convention denotes electrical polarity with the specimen in compression. Specimens oriented from −X to +X give a positive electrical signal in our experimental arrangement, so they will be called +X oriented. Specimens oriented from +X to −X will be called −X oriented. The orientation is further clarified in Fig. 1.

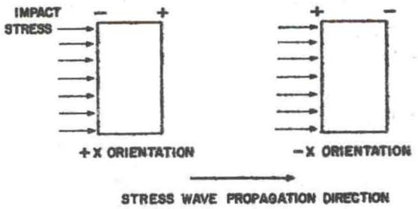

FIG. 1. Orientation of the crystals.

THE NEGATIVE ANOMALY

The results reported previously were taken from specimens with +X orientation. The charge release from these specimens was found to be linear to 25 kbar. A distinctly different behavior is found for −X specimens as illustrated in Fig. 2. From ordinary low signal considerations one would expect the −X specimens to give a charge which is the negative of that obtained for the +X specimens. The low signal behavior is observed for impact stress less than 8 kbar but different behavior is observed for higher stress. Anomalies in the behavior of −X specimens when stressed to about 65 kbar were first noted by Neilson and Benedick.[5] They observed positive charge from −X specimens in contradiction to the low signal behavior.

In changing the x-axis electric polarity relative to the stress propagation direction no mechanical conditions are changed except for coupling from electrical to mechanical which is small for quartz. However, the change in electric polarity does reverse the direction of the field through the stressed portion of the crystal produced by the stress induced polarization. One must conclude that it is the effect of the field reversal which produces the negative anomaly. Since there are no indications of any change in the elastic constant governing the propagation

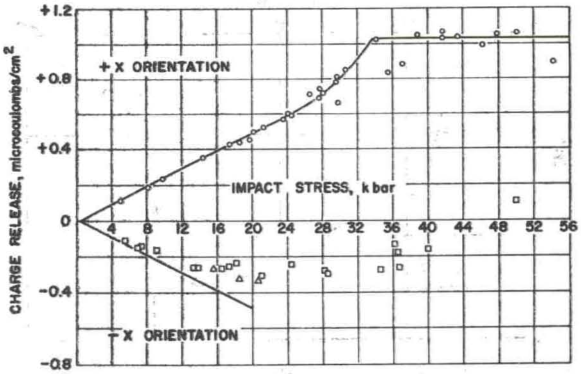

FIG. 2. Charge anomaly for negative orientation.

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* This work was done under the auspices of the U. S. Atomic Energy Commission.

[1] R. A. Graham, J. Appl. Phys. **32**, 555 (1961).

[2] R. A. Graham, Bull. Am. Phys. Soc. **5**, 511 (1960).

[3] For a comprehensive description of the technique see R. A. Graham, Rev. Sci. Instr. **32**, 1308 (1961).

[4] M. H. Rice, R. G. McQueen, and J. M. Walsh in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1958), Vol. VI, p. 8.

[5] F. W. Neilson, and W. B. Benedick, Bull. Am. Phys. Soc. **5**, 511 (1960).

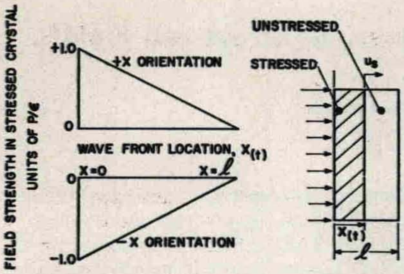

FIG. 3. Field orientation in stressed crystal.

characteristics of the stress wave front, the effects are felt to be largely changes in the electric strength characteristics of the stressed crystal under high electric field and high stress.

Neilson proposed that the anomaly for stress greater than above 50 kbar could be explained in terms of a unilateral conduction mechanism depending on the direction of the electric field. Negative carriers were proposed to be the result of an ionization process occurring at the stress wave front. These carriers, presumably electrons, are then accelerated into the stressed portion of the crystal by the high piezoelectric field. One would expect that the extent of the effect would depend upon the magnitude of the stress and field, both being directly related in a linear piezoelectric medium. In the experiments reported here the magnitude of the transient stress was varied from 5 kbar to about 50 kbar to observe the progressive deterioration of dielectric strength.

If we take a model of an elastic stress wave propagating in a piezoelectric medium under one dimensional, linear mechanical and electrical conditions with short circuit conditions external to the crystal, the field in the stressed portion of the crystal for a particular stress is given by[6]

$$E=(P/\epsilon)[1-(u_st/l)],$$

where E=electric field, P=polarization produced by the application of the stress, ϵ=dielectric permittivity of the stressed crystal, u_s=wave front propagation velocity, t=time reckoned from the instant of impact, and l=thickness between the two electrodes. The crystal model and the resulting field are shown in Fig. 3. Here the maximum field occurs at $t=0$ and is equal to P/ϵ. Using Bechmann's[7] low signal constants for quartz for one-dimensional strain conditions the maximum field is 4.93×10^4 v/cm-kbar. Maximum fields encountered in our experiments and computed on this basis range from 2.5×10^5 v/cm to 2.5×10^6 v/cm. These fields are less than the steady-state breakdown strength for X-cut quartz at atmospheric pressure. Von Hipple and Maurer[8] found this to be 5.8×10^6 v/cm at 25°C.

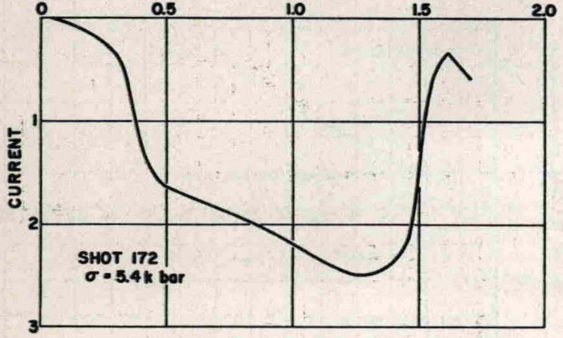

FIG. 4. 5-kbar experimental waveform.

EXPERIMENTAL CURRENT OBSERVATIONS

The characteristics of the electric breakdown process are illustrated in Figs. 4–9 which show the current vs time waveforms obtained in the various stress regions. The figures are normalized in amplitude by the stress so that if no nonlinearity or anomaly occurs they should all appear the same. These waveforms are typical and quite reproducible. Below 8 kbar the smooth undistorted shape in Fig. 4 is obtained. This record is the same as that observed from a +X specimen except that the sign of the current is reversed as expected of normal low signal behavior. Figure 5 shows several distinct periods of partial breakdown when the stress is increased to 9 kbar. With increasing stress up to 25 kbar the waveform of Fig. 6 is typical of the increasing internal conduction without complete disruptive breakdown. The major distortion of the waveform is at the time of maximum field. Between 25 and 34 kbar, Fig. 7 is typical in that the current rises to a negative value then drops to zero in times of about 10^{-7} sec indicating a disruptive breakdown at the time of maximum field. Above 34 kbar, as shown in Fig. 8, the internal conduction following breakdown is much in evidence during the entire transit time of the wave. Figure 9 shows gross positive currents following the disruptive breakdown

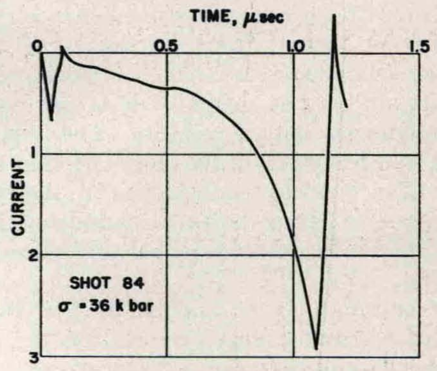

FIG. 5. 36-kbar experimental waveform.

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[6] G. W. Anderson, Colloque International 1961 Sur Les "Ondes de Détonation" Paris, France, August 28 to September 2, 1961; also, Sandia Corporation Reprint SCR-416, June 1961.

[7] R. Bechmann, Phys. Rev. **110**, 1060 (1958).

[8] A. Von Hipple and R. J. Maurer, Phys. Rev. **59**, 820 (1941).

for stress of approximately 52 kbar. This behavior appears to be a continuation of the trend beginning at 34 kbar. Figures 8 and 9 show two distinct processes occurring. The disruptive breakdown is very pronounced in both figures. The later behavior depends on the stress amplitude with either positive currents or internal conduction occurring for the full transit of the wave.

CHARACTERISTICS OF THE BREAKDOWN PROCESS

Several features of the negative anomaly shown should be summarized. The effect begins at low stress levels in regions considered by us to be macroscopically elastic. The field is definitely shown to be essential to the breakdown since even in the case of substantial positive currents the initial instantaneous negative field is shown to be present. There are distinct stresses and/or fields at which a change in behavior is observed. The first indication of a breakdown occurs at a field more than an order of magnitude lower than the steady-state breakdown field. The breakdown observed is definitely in the quartz itself since the specimens are carefully encapsulated in an epoxy potting compound and identical experiments for +X specimens show smooth waveforms with no evidence of breakdown. The dielectric breakdown nature of the phenomenon is also substantiated by high speed camera observations of the internal luminescence of quartz for high stress impulsive loading reported by Brooks and Neilson.[9]

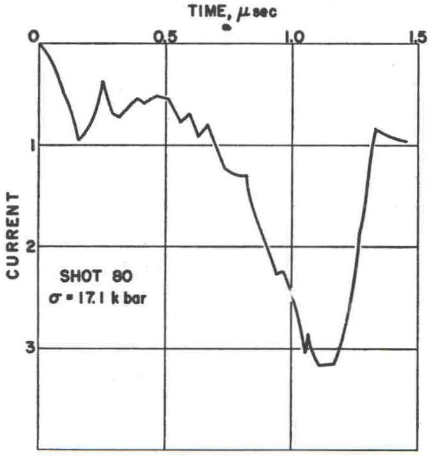

FIG. 6. 17-kbar experimental waveform.

The anomaly is also present for natural quartz. The charge observed for the three experiments performed on natural quartz are identified by the triangles in Fig. 2.

Since the breakdown process begins at such a low electric field, the effect of the transient stress is to lower the electric strength. Under static stress it has been reported[10] that for several materials the electric strength increases with compression until the elastic limit is reached and a sudden decrease in electric strength occurs. Distinct changes in the negative anomaly occur for stresses at which nonlinear mechanical behavior has been observed for quartz. Bridgman performed three linear compressibility experiments on natural quartz.[11–13] In the vicinity of 6 to 8 kbar a change in compressibility was observed. The reported crushing strength for X-cut quartz is 22 kbar.[14] Our +X experiments show a gross discontinuity in the charge-stress relation at 34 kbar. This discontinuity has the appearance of a gross mechanical yield.

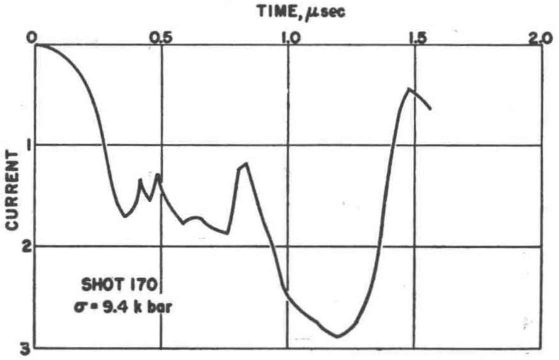

FIG. 7. 9–kbar experimental waveform.

Quartz is known to contain far fewer dislocations than metals and plastic deformation of quartz has not been

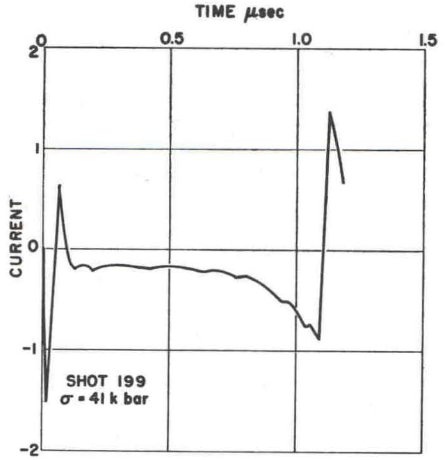

FIG. 8. 41-kbar experimental waveform.

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[9] W. P. Brooks and F. W. Neilson, Bull. Am. Phys. Soc. **5**, 511 (1960).

[10] S. Whitehead, *Dielectric Breakdown of Solids* (Oxford University Press, Cambridge, England, 1953), p. 108.

[11] P. W. Bridgman, Am. J. Sci. **10**, 483 (1925); also shown in R. B. Sosman, *Properties of Silica, Part II* (Book Department, The Chemical Catalog Company, Inc., New York, 1927), p. 430.

[12] P. W. Bridgman, Am. J. Sci. **15**, 287 (1928).

[13] P. W. Bridgman, Proc. Am. Acad. Arts Sci. **77**, 190 (1949).

[14] R. B. Sosman, *International Critical Tables* (McGraw-Hill Book Company, Inc., New York, 1928), Vol. IV, p. 21.

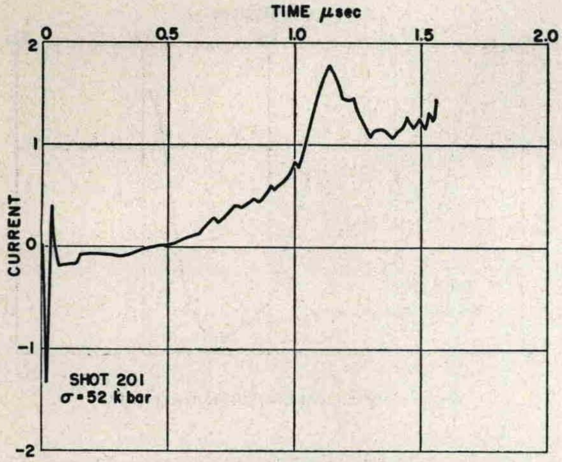

FIG. 9. 52-kbar experimental waveform.

clearly established. Dislocation densities of from 10^2 to 10^3 cm^{-2} have been reported by Hiki[15] for synthetic quartz obtained from the same source as ours.[16]

Since the observed stress for which the dielectric strength changes correlates with mechanical nonlinear stresses, a connection to dislocation motion is suggested. This correlation suggests that the ionization process is a high velocity dislocation motion at the stress wavefront. The observed low stress behavior can be explained if the dislocation motion results in liberated electrons. These experiments are unique in that a slight dislocation motion which might not be observable as plastic deformation can result in profound effects on the electric strength because of the increase in energy due to the high electric field.

Evidence of ionization due to dislocation motion has been shown by Gilman[17] who observed that dislocation motion in LiF results in charge being accumulated on the dislocation. Fischbach and Nowick[18] have reported theoretical and experimental evidence for charge separation due to dislocation motion in NaCl.

SUMMARY

A dielectric breakdown process for −X oriented quartz under transient conditions is observed which may be described as being triggered by stress-induced dislocation motion of various degrees resulting in liberated electrons which are accelerated into the stressed portion of the crystal. For the +X specimens the field direction would keep any liberated electrons at the wavefront, and their influence would only become significant when the charge became large relative to the piezoelectric charge. Once the electrons are accelerated into the stressed crystal with sufficient energy, an electric breakdown process will operate. For low energies, the impaction of the electron produces only partial breakdown, but for high energies an avalanche effect results. Above a stress which is thought to be the mechanical yield, positive currents are observed after the avalanche breakdown.

All the data given in this paper were taken from specimens of one-half inch diameter and one-quarter inch long. Under transient conditions the geometry complicates the later time behavior of the crystal. The times close to impact time, however, are a close approximation to one-dimensional behavior. Although the geometry of the crystal is not expected to alter any of the general features of the negative anomaly a quantitative description of the behavior should be observed on one-dimensional geometry specimens. Experiments on a one-dimensional geometry are planned.

ACKNOWLEDGMENTS

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[15] Y. Hiki, J. Phys. Soc. Japan **16**, 664 (1961).

[16] Our quartz is Y-bar crystal grown hydrothermally by Sawyer Research Products, Inc.

[17] J. J. Gilman, *Fracture*, edited by B. L. Auerbach, D. K. Felbeck, G. T. Hahn, and D. A. Thomas (John Wiley & Sons, Inc., New York, 1959), p. 209.

[18] D. B. Fischbach and A. S. Nowick, J. Phys. Chem. Solids **5**, 302 (1958).

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