

Dielectric Anomaly in Quartz for High Transient Stress and Field*

R. A. GRAHAM

Sandia Corporation, Albuquerque, New Mexico

(Received November 27, 1961)

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 technique³ involves impacting flat-faced cylinders of quartz crystals upon each other with precise control being maintained on alignment. By using the Hugoniot conservation of momentum⁴ 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.

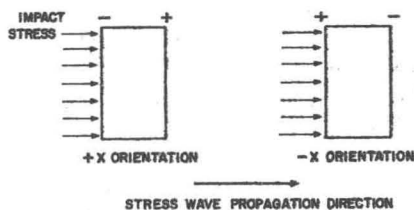


FIG. 1. Orientation of the crystals.

* 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.

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

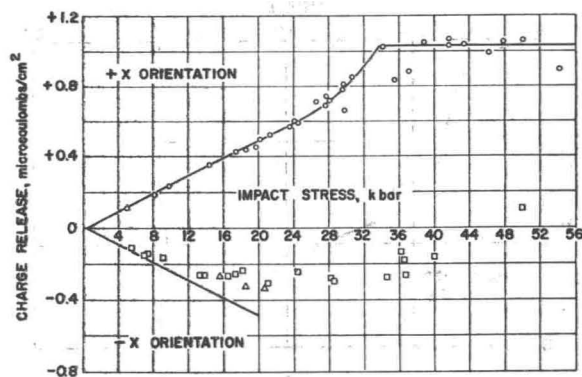


FIG. 2. Charge anomaly for negative orientation.

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

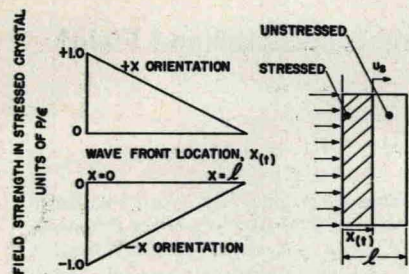


Fig. 3. Field orientation in stressed crystal.

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 by⁶

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

where E = electric field, P = polarization produced by the application of the stress, ϵ = dielectric permittivity

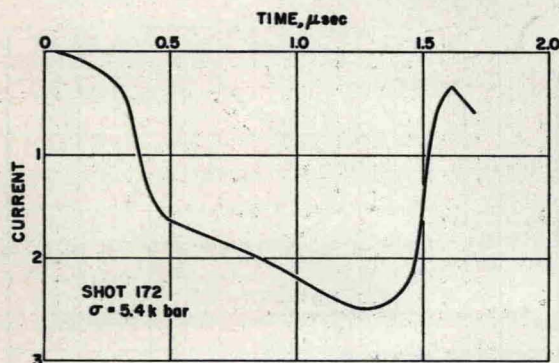


Fig. 4. 5-kbar experimental waveform.

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.

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's⁷ 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 Hippel and Maurer⁸ found this to be 5.8×10^6 v/cm at 25°C.

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

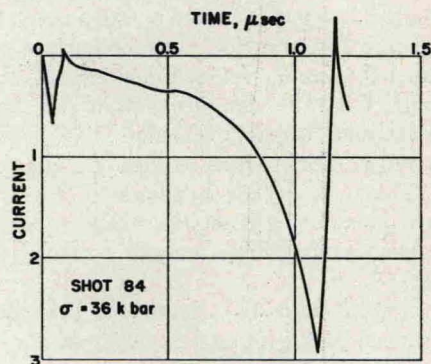


Fig. 5. 36-kbar experimental waveform.

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

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

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