Observations of intense scintillations in unirradiated glycine*

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We report new observations of scintillations and current pulses from glycine, a nonferroelectric crystal, which result from heating or cooling the sample between 77 and 300 K with no previous irradiation. The scintillations and current pulses occur approximately in coincidence and are probably due to a potential difference that exists across the sample as a result of a temperature gradient. Our results are similar to all previous observations of scintillations in ferroelectric crystals.

Yockey and Aseltine[1] have recently reported observations of intense scintillations in potassium dihydrogen phosphate (KDP) resulting from γ irradiation and subsequent heating. In previous works other ferroelectric crystals have also been shown to exhibit intense scintillations during a temperature ramp. Gilliland and Yockey[2] reported observations of scintillations from both γ-irradiated and unirradiated ferroelectric crystals of Rochelle salt and guanidine aluminum sulfate hexahydrate. They noted that the scintillations occurred upon cooling as well as upon warming and suggested that the scintillations were due to triboluminescence produced by the thermal expansion and contraction of the crystal. Similar scintillations were reported by Yockey[3] to occur in the ferroelectric crystals potassium dihydrogen phosphate (KDP) and triglycine sulfate (TGS). Robertson and Baily[4] found that virgin crystals of TGS would emit light when heated through the ferroelectric Curie temperature and that the light came from electrical discharges into the surrounding atmosphere. A study of electrical sparks from ferroelectric TGS crystals was conducted by Schmidt and Petersson.[5] Their results indicated that electrical discharges into the surrounding atmosphere occurred when the TGS crystal was heated through its Curie point. The roughness of the crystal surface and the configuration of the domains had a strong influence on the effect. Zerem and Halperin[6] found that short-duration light pulses were accompanied by current pulses when x-irradiated KDP was warmed from 77 K. An ambient gas pressure above about 0.1 Torr was required for the pulses to occur.

The comprehensive investigation of scintillations conducted by Yockey and Aseltine[1] culminated in the promulgation of a theory to explain their observations. They proposed a model based upon the strong internal fields in ferroelectrics due to the polarization which exists when the crystal is in the ferroelectric phase to explain the scintillations. They found that it was necessary for the crystal to be in the ferroelectric phase during irradiation and concluded that scintillations are found only in ferroelectrics and that the effect is peculiar to such substances.

In this paper we report new observations of scintillations from glycine, a nonferroelectric crystal,[7] which result from heating or cooling the sample between 77 and 300 K with no previous irradiation. The scintillations probably occur as a result of a voltage being developed between two surfaces of the crystal and their subsequent discharge. The surface charging is originally generated by a change in the temperature of the crystal and the potential difference is believed to be due to the temperature gradient that exists across the sample. Current pulses accompanied the scintillations and the direction of the measured current flow was dependent upon the direction of the temperature gradient.

Experimental measurements were made on single crystals of glycine and deuterated glycine from 77 to 300 K. The samples were placed in an optical Dewar and evacuated to a pressure of 10^{-3} Torr by a glass mercury-vapor-diffusion system. Following evacuation, the samples were cooled at a nonlinear rate to 77 K and subsequently heated to 300 K at a linear rate of 20 K/min. The scintillations were detected by an EMI 9656R photomultiplier tube (S-11 response) and plotted on an X-Y recorder as a function of temperature. Visual observations as well as photographs of the scintillations were made by employing an image intensifier and appropriate optical lenses. The current pulses were measured using the metal strip as one electrode and the copper block as the other electrode (see Fig. 1) and detecting the current with a picoammeter.

Figure 2 shows the observed scintillations that occur in a single crystal of unirradiated glycine as the sample temperature is lowered from 300 to 77 K. The scintillations appear to be superimposed on a thermoluminescence-glow curve, but since these data were taken as the sample cooled,

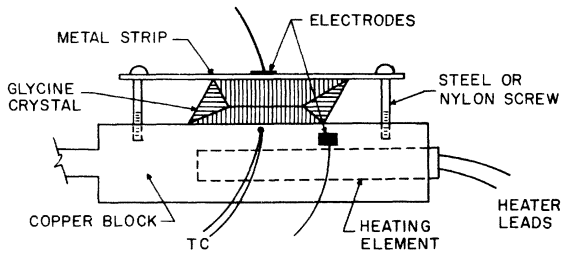

FIG. 1. Sample holder used for making current measurements and observing scintillations. The heating element is a 30-W cartridge resistor inserted into a copper block which is mounted to the cold finger inside the optical Dewar.

we conclude that the peak is not due to thermoluminescence. Repeated measurements of this phenomenon indicate that although the temperature at which a scintillation occurs cannot be predicted; they never occur above 300 K. Several times we heated the sample to 500 K without observing any scintillations above this cutoff temperature. A lower limit on the scintillation activity could not be established since we were still observing the pulses at our lowest operating temperature of 77 K. Replacement of the glycine sample by a deuterated one lowered the cutoff temperature to 286 K, but again we could not establish a lower limit for the scintillation activity.

A vacuum environment was required for the scintillations to occur. If the Dewar attained atmospheric pressure, the scintillations were no longer detected, but they could be completely rejuvenated by reevacuating the Dewar and heating or cooling the sample. We also found that annealing the crystal at 500 K for 1 h did not reduce the number and intensity of scintillations observed in subsequent experiments. In an attempt to determine whether the scintillations were indeed temperature dependent or perhaps time dependent, we maintained the crystal temperature at a constant value for several minutes and observed no scintillations or current pulses.

Visual inspection of the scintillations showed that they began as small discharges and increased in frequency of occurrence until a large scintillation resulted. Photographs revealed that the discharge was along the metal strip used to hold the crystal onto the copper heating block (see Fig. 1). When the steel screws used in mounting the crystal were replaced by nylon ones we observed only a few scintillations. Electrodes were placed on the metal strip and copper heating block as shown in Fig. 1 and current pulses (~nA) were measured as the sample was heated or cooled. For these measurements the nylon screws were used to secure the crystal onto the heating block. We found that the direction of the current flow depended upon the direction of the temperature gradient. As the sample began to cool from room temperature to 77 K, a small current (~pA) began to flow from the higher-temperature crystal surface to the lower one. This current became constant after the crystal temperature had been lowered approximately 30 K. The observed current pulses (~nA) were superimposed upon this constant background current and indicated a flow of charge from the lower-temperature surface of the crystal to the higher one. A reversal of the direction of the temperature gradient caused the measured current flow to change its direction.

These experimental results indicate that surface charging occurs in unirradiated glycine due to a change in the crystal temperature. Presumably, a potential difference is developed between two surfaces of the crystal due to the fact that a temperature gradient exists across the sample, thereby producing more charge on one crystal surface than on the other. Only a few scintillations were detected when the conducting steel screws were replaced by the nonconducting nylon ones. This was because the upper surface of the crystal was insulated from the lower one and surface to sur-

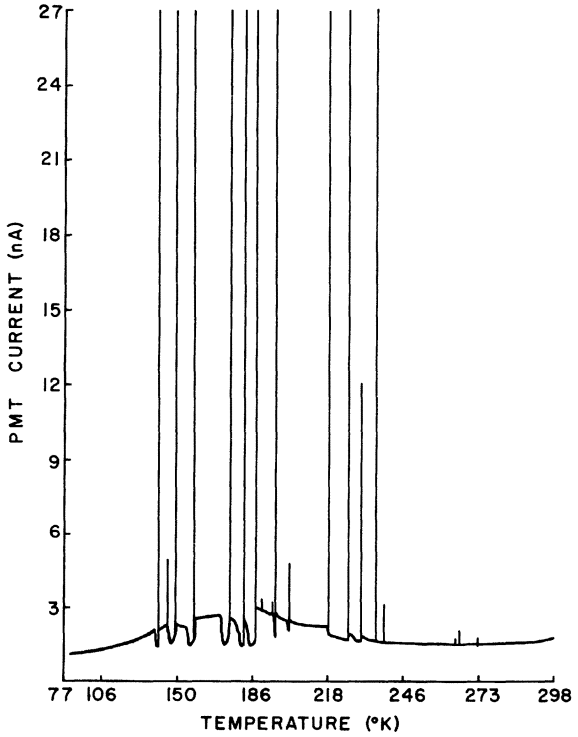

FIG. 2. Scintillations observed from glycine by cooling the sample from 300 to 77 K with no previous irradiation.

face discharging could not take place. The current pulses were a result of the surface to surface discharging through a Keithley picoammeter. The reversal of the current flow due to the pulses is readily explained by the change in the temperature gradient. The sample thickness of 6 mm is sufficient to produce a temperature gradient of 10 to 15 K. When the sample is being cooled, the lower surface of the crystal is at a lower temperature than the upper one. Upon reaching 77 K the crystal is then warmed and consequently the lower surface will be at a higher temperature than the upper one. Since the surface charge is produced by a change in the temperature, each surface acquires charge but at a different rate due to the fact that one surface is at a different temperature. This would also explain our observations of cessation of scintillations when the temperature remained constant.

Our results on unirradiated glycine are very similar to all previous observations of scintillations in ferroelectric crystals. The model proposed by Yockey and Aseltine[1] to explain the scintillation phenomenon in ferroelectrics implied that charge liberated as a result of ionizing radiation was trapped at domain walls. These sites are unique to ferroelectrics and are required to be present during irradiation. The potential difference developed depended upon the release of this trapped charge and the temperature differential $T - T_0$, where T is the crystal temperature and T_0 is the Curie point. The data presented in this paper also suggests a potential difference being developed across the crystal as a result of a temperature gradient, but it cannot depend upon a Curie temperature. Finally, we have made similar observations of scintillations from pressed pellets of powdered glycine, and thereby conclude that a large single crystal is not required for the phenomenon to manifest itself.

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