(ii) The modulus-compensated activation energy for creep in the temperature range 650–750 °C is 2.09 ± 0.08 eV (48.17 ± 1.84 kcal/mole), suggesting that the creep rate is controlled by the lattice diffusion of the fluorine ion.

(iii) The creep strain rate is proportional to (σ)n, with n = 5.11 for T = 650 and 700 °C and n = 3.74 for T = 750 °C.

(iv) At the temperatures and stresses employed in the present study, the creep rate is controlled by dislocation motion through the lattice.

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Pyroelectric Voltage Response to Rectangular Infrared Signals in Triglycine Sulphate and Strontium-Barium Niobate

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The pyroelectric voltage response to rectangular signals of infrared radiation is derived using our recent analysis of the response to a step signal. The response is symmetrical with respect to the electronic and thermal time constants, τ_e and τ_T of the samples, and with respect to the times T_1 and T_2 of irradiation and darkening. When τ_e differs appreciably from τ_T, the rise of the response at the onset of irradiation and its fall at the beginning of darkening are exponential with the same time constant, which is the smaller one from among τ_e and τ_T. A rectangular response is obtained when τ_e ≪ (T_1 / T_2) ≪ τ_T. In this case the peak-to-peak value V_{pp} of the response is proportional to τ_e and independent of the pulse frequency, as well as of the ratio T_1 / T_2. When T_1 and T_2 are each less than the smaller one from among τ_e and τ_T, triangular responses are obtained, for which V_{pp} is independent of τ_e and inversely proportional to the pulse frequency. Pyroelectric responses obtained in triglycine sulphate and strontium-barium niobate samples of different τ_T were found to follow the derived expression, and plots of the measured parameters of the response vs τ_T, T_1 / T_2, and pulse frequency show good agreement with the analysis.

I. INTRODUCTION

The response of a pyroelectric detector differs substantially from that of other radiation detectors. It starts almost simultaneously with any change in irradiation, but when the irradiation is already constant, the pyroelectric response continues to rise, peaks, and decreases to zero.

The pyroelectric response was described by Chynoweth* and Cooper.7 White5 has analyzed the voltage response to a step signal and the current response to square-wave modulated excitation. Various response shapes observed with square and sinusoidal modulation were analyzed in NASA reports.4 A quite comprehensive treatment of the response, especially to sinusoidal signals, was given by Hadni.5

In a previous paper8 we reported on the pyroelectric voltage response to a step infrared radiation signal.

The analytically derived parameters of the response agreed well with experiment in detectors made of triglycine sulphate (TGS) and strontium-barium niobate (SBN). The response is symmetric with respect to the electronic and thermal time constants of the samples. The initial slope of the response, divided by the absorbed radiation flux, was found to be a sample constant and, under certain conditions, even a material constant.

These results are used in the present paper for a study of the pyroelectric voltage response to rectangular radiation pulses. Analytical expressions are derived for the parameters of the response as functions of the signal parameters and of the thermal and electronic time constants of the detector. These expressions are compared with experiment in TGS and SBN samples.

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II. EXPERIMENTAL TECHNIQUE

The samples were thin (10–40 μm) slices of TGS or SBN, electroded on their major faces perpendicular to the polar axis and mounted in vacuum in transistor cases with infrared transparent windows. The radiation source was a 500 °K blackbody and a variable speed modulator (5–40 rps) with choppers having different on/off time ratios. To provide rectangular pulses, the width of the chopper teeth, as well as the distance between them, were much larger than the diameter of the radiation beam. The measuring circuit consisted of interchangeable load resistors (10⁶–10¹² Ω) connected parallel to the sample and to a FET source-follower circuit. The obtained pyroelectric response voltages were measured on a Tektronix CRO with a 3A9 amplifier.

III. SYMBOLS OF THE STEP-SIGNAL RESPONSE (REF. 6)

\[ A \] electrod sample area;
\[ C \] capacitance of sample and circuitry at preamplifier input;
\[ C_T \] thermal capacity of sample;
\[ F_0 \] flux absorbed in sample during a step radiation signal, W/cm²;
\[ G_T \] thermal conductance of sample to ambient;
\[ k = \xi F_0 \] initial slope of the voltage response, V/sec;
\[ R \] resistance of sample and circuitry at preamplifier input;
\[ \theta = \tau_e/\tau_T \]
\[ \lambda \] pyroelectric coefficient, C/cm² deg;
\[ \xi = \lambda A^2/CC_T \] sample (or material) constant, cm²/C;
\[ \tau_e = RC \] electronic time constant of sample and circuitry at preamplifier input;
\[ \tau_T = C_T/G_T \] thermal time constant of sample.

IV. THEORETICAL ANALYSIS

A train of \( N \) rectangular radiation pulses of width \( T_s \), height \( F_o \), and separation \( T_d \) from each other can be represented by a sum \( F_n(t) \) of \( 2N \) displaced step functions \( F(t) \), whose heights are \( F_o \) for positive arguments and zero otherwise; hence

\[ F_n(t) = \sum_{n=0}^{N-1} \left[ F(t-nT) - F(t-nT - T_s) \right], \]  

where \( T = T_s + T_d \) is the pulse period. We assume that the radiation is absorbed uniformly throughout the pyroelectric crystal, causing a small temperature rise. The temperature of the crystal is assumed far below the Curie point. Due to the linearity of the pyroelectric voltage response as a function of the excitation, the response \( V_n(t) \) to \( F_n(t) \) is

\[ V_n(t) = \sum_{n=0}^{N-1} \left[ V(t-nT) - V(t-nT - T_s) \right]. \]  

The step-signal responses \( V(t) \) are given by

\[ V(t) = V_o \exp(-t/\tau_e) - \exp(-t/\tau_T) \]  

for positive arguments and are zero otherwise; here \( V_o = k \tau_o/(1-\theta), \theta \neq 1 \).

The derivation of the response to a pulse from the response to displaced step signals is explained graphically in Fig. 1 for the case of a single pulse (\( N = 1 \)). Until the end of the pulse the response is identical to the step-signal response. After the end of the radiation pulse, the response decays to a minimum value below the time axis, then returns to zero. When \( \tau_T \) differs appreciably from \( \tau_e \), the decay of the response to the minimum value and its subsequent return to zero are exponentials whose time constants are, respectively, the smaller and the larger one from among \( \tau_e \) and \( \tau_T \).

![Fig. 1. Pyroelectric voltage response \( V_n(t) \) to a single rectangular radiation pulse \( F_n(t) \) of height \( F_o \) and duration \( T_s \). Dotted curves show the responses \( V(t) \) and \( -V(t-T_s) \) to radiation-on and radiation-off step signals, \( F(t) \) and \( -F(t-T) \), respectively.](image)

![Fig. 2. Steady-state pyroelectric voltage responses to rectangular radiation pulses of different frequencies in TGS sample F-S. F_o = 1 mW/cm²; \( T_s = T_d \); \( R_L = 55 \Omega \).](image)
The response to the first pulse of a train is obviously identical to the response to a single pulse during \( t < T \). It can be shown that the response to subsequent radiation pulses in a train shifts down with respect to the time axis and changes shape until the areas enveloped by the response above and below the time axis during one cycle become equal. \(^7\) Thereafter, the responses to each subsequent radiation pulse are identical. This steady state is reached at a time longer than the larger one from among \( \tau_\varepsilon \) and \( \tau_r \).

To analyze the response to a radiation pulse of the train in the steady state we start counting a time \( t'\) from the beginning of this pulse \( 0 < t' < T_1 \). At the end of the radiation pulse we start counting a time \( t''\) \( (0 < t'' < T_2) \). For these intervals, we obtain

\[
V_p(t') = V_0 [a_{2T} \exp(-t'/\tau_r) - a_{2T} \exp(-t'/\tau_\varepsilon)],
\]

\[ V_p(t'') = V_0 [a_{2T} \exp(-t''/\tau_r) - a_{2T} \exp(-t''/\tau_\varepsilon)], \tag{4} \]

where the coefficients \( a_{ij} \), \( i = 1, 2 \), \( j = \varepsilon, r \), are given by

\[
a_{ij} = \left[ 1 - \exp(-T_j/\tau_j) \right] \left[ 1 - \exp(-T_j/\tau_j) \right]^{-1}.
\]

The peak value \( V_p^* \) of \( V_p(t') \),

\[
V_p^* = k \tau_r a_{2T} (a_{2T}/a_{2T})^{1/12} \left(1 - \theta\right),
\]

is reached at a time

\[
t_p^* = \tau_r \ln(a_{2T}/a_{2T})(1 - \theta)^{-1}.
\]

The corresponding expressions for \( t_p^* \) and \( -V_p^* \) are obtained by changing the indices 2 to 1.

It is seen that the response is antisymmetric with respect to \( T_1 \) and \( T_2 \), i.e., that interchanging the times of irradiation and darkening makes the response curve flip around the time axis with shape and peak-to-peak value \( V_{pp} \) unaltered. Maximum \( V_{pp} \) is obtained when \( T_1 = T_2 \). The response to a single pulse is obtained by substituting \( T_2 = \infty \) in the coefficients \( a_{ij} \).

Rectangularly shaped responses \([\text{Figs. 2(a)} \text{and} \text{3(i)}-\text{3(l)}]\) are obtained when \( T_1 = T_2 \), where the coefficients \( \tau_e = \tau_r \). The corresponding expressions for \( V_p(t') \) are obtained when \( T_1 = T_2 \).

During a radiation pulse it rises from the minimum value \(-kT_1T_2/2T\) at \( t' = 0 \), to a maximum of \(+kT_1T_2/2T\) at \( t' = T_1 \) crossing the time axis at \( t' = \frac{1}{2} T_1 \). The slope of the rise is \( k^* = kT_2/T \). During the darkening, the response decreases from maximum to minimum with a slope \( k^* = -kT_2/T \), crossing the time axis at \( t' = \frac{1}{2} T_2 \) and \( V_{pp} \) is \( kT_2/T \). When \( T_1 = T_2 \), \( V(t') \) and \( V(t'') \) are symmetrical, \( k^* = -k^* \), and \( V_{pp} = 2kT_2/T \).

![FIG. 3. Steady-state pyroelectric voltage responses to rectangular radiation pulses of different \( T_1/T_2 \) ratios in YGS sample F-8 for various \( \tau_\varepsilon \); \( F_2 = 1 \text{ mW/cm}^2; C = 32 \text{ pF}; \tau_\varepsilon = 2.4 \text{ sec;} T = T_1 + T_2 = 0.1 \text{ sec} \). (a)-(d) \( R_2 = 10^7 \Omega; \tau_\varepsilon = 0.32 \text{ msec} \); (e)-(g) \( R_2 = 10^8 \Omega; \tau_\varepsilon = 3.2 \text{ msec} \); (h)-(l) \( \tau_\varepsilon = 0.32 \text{ sec} \).](image)

![FIG. 4. Peak-to-peak values of the voltage response to rectangular radiation pulses as a function of their period, for different \( \theta \) ratios and \( T_1 + T_2 \). Crosses are at points where \( T_1 = T_2 \).](image)

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The computed dependence of $V_{pe}$ on the period of the pulses in steady state when $T_1 = T_2$ is shown graphically in Fig. 4 for different $\theta$ ratios. On the curves for each $\theta$, there is a cross marking the point where $T_1 = T_2$. To the left of the cross (smaller $T$) is the region of triangular pulses, for which $V_{pe}$ is proportional to $T$; to the right (on curves for $\theta \approx 1$) is the region of rectangular pulses, for which $V_{pe}$ is independent of $T$. With $T$ still increasing, $V_{pe}$ starts rising slowly until (at $T = 10T_0$) it reaches a maximum of twice the peak value of the step-signal response. Here the response to each radiation pulse does not "remember" the previous pulse, therefore it is identical to a doubled step-signal response. For $\theta > 1$, the curves of the figure are still valid if $\tau_s$ and $\tau_e$ are interchanged.

V. EXPERIMENTAL RESULTS

Detailed studies of the pyroelectric response have been carried out at room temperature on 15 TGS samples, $^8$ and on six samples of SBN. $^9$ A typical voltage response to a train of rectangular infrared signals from a 500 K blackbody source is shown in Fig. 2. The oscillogram traces here were obtained on a TGS sample (F-8) having $F_0 = 1$ mW/cm$^2$, and the measured value $b$ of the initial slope is 5 V/sec. The ratio of radiation on/off times is 1/1, and the vertical scale is 1 mV/cm.

The upper trace (Fig. 2(a), time scale 20 msec/cm) was obtained with a chopping frequency 9.3 Hz, so that $\tau_e < T_1 = T_2 = 54$ msec $< \tau_s$, and the response is seen to be close to rectangular. On the lower trace (Fig. 2(b), chopping frequency 59 Hz, and $T_1 = T_2 = 1.3$ msec) the exponential rise and decay of the response last for more than half the pulse duration. However, because $T_1 = T_2$, $V_{pe}$ is still the same as in the foregoing case. From the picture, one can estimate that $V_{pe}$ will remain frequency independent up to $\sim 70$ Hz. The lower trace (Fig. 2(c)) shows that the chopping frequency is 380 Hz. $T_1 = T_2 = 1.3$ msec is slightly smaller than $\tau_s$, and the response is already close to triangular. $V_{pe}$ is smaller, and it is seen that it will decrease with increasing frequency.

The dependence of the response on the ratio $T_1/T_2$ is shown in Fig. 3. The CRO traces here were obtained in sample F-8 under a constant chopping frequency of 10 Hz and $T_1/T_2$ ratios varying from 1/1 (upper row) down to 1/4, 1/10, and 1/23. The rectangular responses (traces a-d, 1 mV/cm on vertical scale) were obtained with $R_L = 10^8 \Omega$, so that $\tau_e = 0.32$ msec, and the condition $\tau_e < T_1, T_2$ is satisfied. $V_{pe}$ is seen to be independent of $T_1/T_2$ and equal 1.5 mV, as required by the formula $V_{pe} = k\tau_e$. The traces of the middle column (traces e-h, 5 mV/cm vertical scale) were obtained with $R_L = 10^9 \Omega$, $\tau_e = 3.2$ msec. On the two upper traces $T_1$ exceeds $\tau_e$, and even $T_2$; $V_{pe}$ is 16 mV, as for rectangular pulses, and does not depend on $T_1/T_2$.

With $T_1/T_2 = 0.1$ (trace g) $T_1$ is still larger than $\tau_e$, but smaller than $T_2$. However, due to the flatness of the top of the response, $V_{pe}$ is still close to the "rectangular" value. With $T_1/T_2 = 1/23$ (trace h) the response is being cut during its exponential rise and is close to triangular.

The triangular responses (traces 1-4, vertical amplification 50 mV/cm) were obtained with $R_L = 10^9 \Omega$ to make $\tau_e = 0.32$ sec $< T_1, T_2$. It is seen that the response rises linearly during $T_1$, and falls linearly during $T_2$. With decreasing $T_1/T_2$ the slope $k^{'\prime}$ of the rise increases, the slope $k^\prime$ of the fall decreases (in absolute value), and so does $V_{pe}$. The dependence of $V_{pe}$ on $T_1/T_2$ is shown in Fig. 5 for three samples, seeming to fit well the analytical curves.

On each close to rectangular response the rise and fall are both exponentials with a time constant equal to $\tau_e$. The switching of this time constant to $\tau_s$ when $\tau_e$ exceeds $\tau_s$ is shown in Fig. 6. The two traces here represent the pyroelectric responses in a SBN sample (B-1) of $C = 120$ pF and $\tau_e = 0.7$ sec; $F_0 = 0.5$ mW/cm$^2$. Trace a (20 msec/cm horizontally, and 1 mV/cm vertically) was obtained with $R_L = 10^8 \Omega$, so that $\tau_e$
Structure and Composition of Sputtered Tantalum Thin Films on Silicon Studied by Nuclear and X-Ray Analysis

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Microanalysis by direct observation of nuclear reactions and by backscattering of He ions was used to determine the composition of sputtered Ta films on silicon. Oxygen and nitrogen concentration as low as 0.3 and 0.1%, respectively, were found for bcc and β-Ta structures.

At a low deposit rate, oxygen incorporation in β-Ta structure up to about 20% was observed and correlated to a resistivity increase. The Ar/Ta ratio varies between 1.5 and 3% with increasing sputtering power.

INTRODUCTION

The physical and electrical properties of thin films often differ from those of the bulk material. This effect is marked in the case of Ta. Thus, depending upon the deposition technique used, the structure of the Ta obtained may be fcc, bcc, tetragonal, or amorphous. The essential reason for the preferential creation of one or the other structure is not clearly defined in the literature. Some authors tried to establish a relation between structure of the films and a possible incorporation of foreign atoms (oxygen, nitrogen, carbon, etc.) during deposition; others related structures to deposition parameters (temperature, rate, etc.). When considering only the diode sputtering technique, all publications since 1964 agree on the formation of two crystalline forms: bcc (α-Ta) and tetragonal (β-Ta); the system is associated with a higher resistivity and a TCR near zero; its occurrence could be related to the purity of the deposit, the temperature, or the rate of deposition. Read and Altman did not find any marked difference between the impurity content of β and α structures. Sosnak et al. found that formation of β-Ta does not depend on the gas background in argon glow discharge. Gerstengel and Callbrick, Krikorian and Sneed, McLean, and Baker observed that for defined sputtering conditions that normally produce the α form, the increase of oxygen or nitrogen concentration in the discharge atmosphere always produces the α phase. Later on, Westwood and Westwood and Livermore maintained that an increase of oxygen partial pressure in the discharge is associated with the forma-