AIP | Journal of Applied Physics

Pyroelectric Voltage Response to Rectangular Infrared Signals in Triglycine Sulphate and StrontiumBarium Niobate

A. Shaulov and M. Simhony

Citation: J. Appl. Phys. **43**, 1440 (1972); doi: 10.1063/1.1661338
View online: http://dx.doi.org/10.1063/1.1661338
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v43/i4
Published by the American Institute of Physics.

(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 $(\sigma)^n$, 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.

ACKNOWLEDGMENTS

The authors would like to express their appreciation to Robert Terry and Richard Lincoln for many helpful suggestions. The support of this work by the United States Atomic Energy Commission is gratefully acknowledged.

[1]P. M. Burke and O. D. Sherby in *Progress in Materials Science*, edited by B. Chalmers and W. Hume-Rothery (Pergamon, New York, 1967), Vol. 13, No. 7.
[2]F. Schuh, W. Blum, and B. Ilschner, Proc. Brit. Ceram. Soc. 15, 143 (1970).
[3]D. R. Cropper and T. G. Langdon, Phil. Mag. 18, 1181 (1968).
[4]J. Nadeau, Ph.D. thesis (University of California, Berkeley, California, 1960) (unpublished).
[5]J. J. Gilman and W. G. Johnston, in *Solid State Physics*, edited by D. Turnbull and F. Seitz (Academic, New York, 1962).
[6]Y. Haven, Rec. Trav. Chem. Pays-Bas, 69, 1471 (1950).
[7]E. Barsis, E. Lilley, and A. Taylor, Proc. Brit. Ceram. Soc. 9, 203 (1967).
[8]T. G. Stoebe and P. L. Pratt, Proc. Brit. Ceram. Soc. 9, 181 (1967).
[9]F. R. N. Nabarro, *Report of a Conference on the Strength of Solids* (The Physical Society, London, 1948), p. 45.
[10]J. Weertman, J. Appl. Phys. 26, 1213 (1955); 28, 362 (1957).
[11]C. R. Barrett and W. D. Nix, Acta Met. 13, 1247 (1965).
[12]J. E. Dorn, in *Modern Chemistry for the Engineer and Scientist*, edited by W. D. Robertson (McGraw-Hill, New York, 1956), p. 276.
[13]Yu. M. Chernov and A. V. Stepanov, Sov. Phys. Solid State 3, 2097 (1962).
[14]C. B. Pierce, Phys. Rev. 123, 744 (1961).
[15]M. Eisenstadt, Phys. Rev. 132, 630 (1963).
[16]T. G. Stoebe and R. A. Huggins, J. Mater. Sci. 1, 117 (1966).

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

A. Shaulov* and M. Simhony
The Racah Institute of Physics, The Hebrew University, Jerusalem, Israel
(Received 26 May 1971)

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 exponentials with the same time constant, which is the smaller one from among τ_e and τ_T. A rectangular response is obtained when $\tau_e \ll (T_1, T_2) \ll \tau_T$. In this case the peak-to-peak value $V_{\rm ptp}$ 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 $V_{\rm ptp}$ 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_e, 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[1] and Cooper.[2] White[3] 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 paper[6] 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.

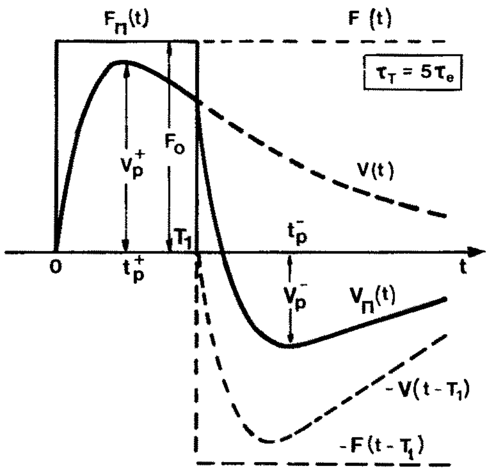

FIG. 1. Pyroelectric voltage response $V_\Pi(t)$ to a single rectangular radiation pulse $F_\Pi(t)$ of height F_0 and duration T_1. Dotted curves show the responses $V(t)$ and $-V(t-T_1)$ to radiation-on and radiation-off step signals, $F(t)$ and $-F(t-T)$, respectively.

II. EXPERIMENTAL TECHNIQUE

The samples[6] 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 (10^6–10^{12} Ω) connected parallel to the sample and to a FET source-follower circuit.[6] The obtained pyroelectric response voltages were measured on a Tektronix RM564 CRO with a 3A9 amplifier.

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

A	electroded 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/cm^2;
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;
θ	$=\tau_e/\tau_T$;
λ	pyroelectric coefficient, C/cm^2 deg;
$\xi\ =\lambda A^2/CC_T$	sample (or material) constant, cm^2/C;
τ_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_1, height F_0, and separation T_2 from each other can be represented by a sum $F_\Pi(t)$ of $2N$ displaced step functions $F(t)$, whose heights are F_0 for positive arguments and zero otherwise; hence

$$F_\Pi(t)=\sum_{n=0}^{N-1}[F(t-nT)-F(t-nT-T_1)], \qquad (1)$$

where $T=T_1+T_2$ 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 $V_\Pi(t)$ to $F_\Pi(t)$ is

$$V_\Pi(t)=\sum_{n=0}^{N-1}[V(t-nT)-V(t-nT-T_1)]. \qquad (2)$$

The step-signal responses $V(t)$ are given[6] by

$$V(t)=V_0[\exp(-t/\tau_T)-\exp(-t/\tau_e)] \qquad (3)$$

for positive arguments and are zero otherwise; here $V_0=k\tau_e/(1-\theta)$, $\theta\neq 1$.

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 this 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 τ_e differs appreciably from τ_T, 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 τ_e and τ_T.

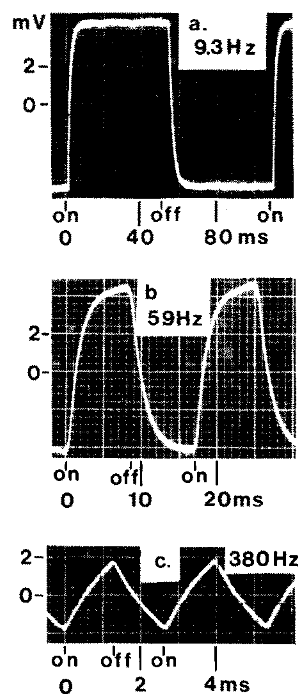

FIG. 2. Steady-state pyroelectric voltage responses to rectangular radiation pulses of different frequencies in TGS sample F-8. F_0 = 1 mW/cm^2; $T_1=T_2$; R_L = 55 MΩ; C = 32 pF; τ_T = 2.4 sec; τ_e = 1.8 msec.

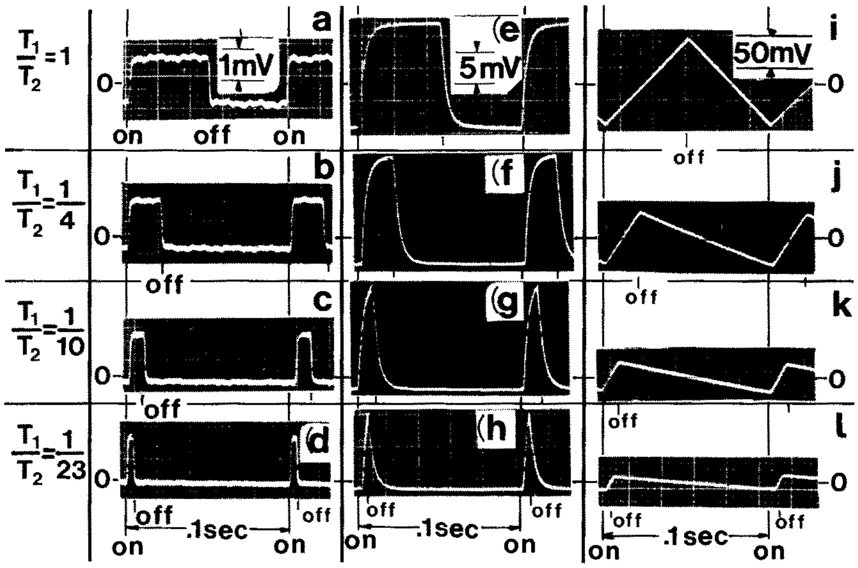

FIG. 3. Steady-state pyroelectric voltage responses to rectangular radiation pulses of different T_1/T_2 ratios in TGS sample F-8 for various τ_e. $F_0=1$ mW/cm^2; $C=32$ pF; $\tau_T=2.4$ sec; $T=T_1+T_2=0.1$ sec. (a)–(d) $R_L=10^7$ Ω, $\tau_e=0.32$ msec; (e)–(h) $R_L=10^8$ Ω, $\tau_e=3.2$ msec; (i)–(l) $R_L=10^{10}$ Ω, $\tau_e=0.32$ sec.

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 τ_e and τ_T.

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

$$V_{\text{II}}(t^+)=V_0[a_{2T}\exp(-t^+/\tau_T)-a_{2e}\exp(-t^+/\tau_e)],$$
$$V_{\text{II}}(t^-)=-V_0[a_{1T}\exp(-t^-/\tau_T)-a_{1e}\exp(-t^-/\tau_e)], \qquad (4)$$

where the coefficients a_{ij}, $i=1, 2$, $j=e, T$, are given by

$$a_{ij}=[1-\exp(-T_i/\tau_j)][1-\exp(-T/\tau_j)]^{-1}.$$

The peak value V_p^+ of $V_{\text{II}}(t^+)$,

$$V_p^+=k\tau_T a_{2e}(\theta a_{2T}/a_{2e})^{1/(1-\theta)},$$

is reached at a time

$$t_p^+=\tau_e\ln(a_{2e}/\theta a_{2T})(1-\theta)^{-1}.$$

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_{ptp} unaltered. Maximum V_{ptp} 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 [Figs. 2(a) and 3(a)–3(d)] are obtained when $\tau_e \ll (T_1, T_2) \ll \tau_T$. In this case, the response to each radiation pulse rises from a value close to $V_p^-=-k\tau_e T_1/T$ with an initial slope $k^+=k$ and reaches $V_p^+=k\tau_e T_2/T$ almost exponentially with a time constant τ_e. At the end of the radiation pulse the response decays from a value close to V_p^+ with an initial slope $k^-=-k$, and reaches V_p^- almost exponentially with the time constant τ_e. For rectangular pulses $V_{\text{ptp}}=k\tau_e$, independent of the ratio T_1/T_2.

Nearly triangular responses [Figs. 2(c) and 3(i)–3(l)] are obtained when T_1 and T_2 are each less than the smaller one from among τ_e and τ_T. In this case

$$V(t^+)=kT_2(t^+-\tfrac{1}{2}T_1)/T,$$
$$V(t^-)=-kT_1(t^--\tfrac{1}{2}T_2)/T.$$

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 $k^-=-kT_1/T$, crossing the time axis at $t^-=\frac{1}{2}T_2$, and V_{ptp} is kT_1T_2/T. When $T_1=T_2$, $V(t^+)$ and $V(t^-)$ are symmetrical, $k^+=-k^-=\frac{1}{2}k$, and $V_{\text{ptp}}=\frac{1}{4}kT$.

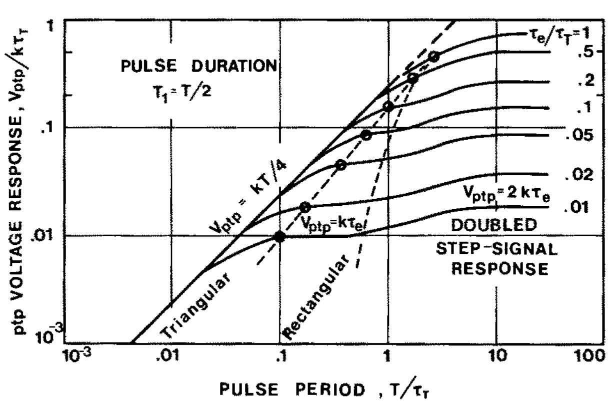

FIG. 4. Peak-to-peak values of the voltage response to rectangular radiation pulses as a function of their period, for different θ ratios and $T_1=T_2$. Crosses are at points where $T_1=t_p^+$.

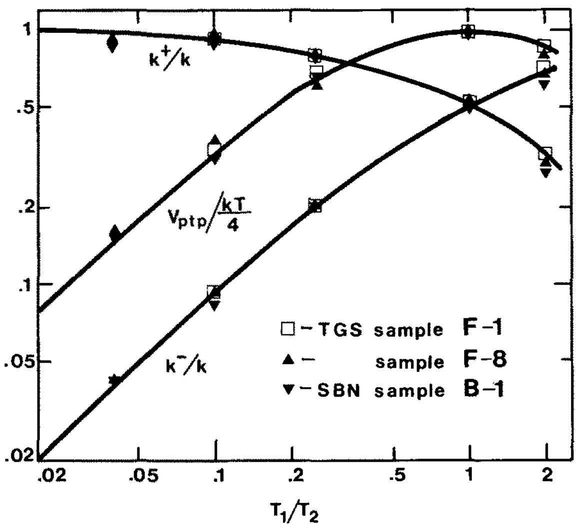

FIG. 5. Peak-to-peak values and slopes of the triangular response to rectangular signals as functions of T_1/T_2 (normalized). Curves are analytical, and points experimental.

The computed dependence of V_{ptp} on the period of the pulses in steady state when $T_1 = T_2$ is shown graphically in Fig. 4 for different θ ratios. On the curves for each θ, there is a cross marking the point where $T_1 = t_p^+$. To the left of the cross (smaller T) is the region of triangular pulses, for which V_{ptp} is proportional to T; to the right (on curves for $\theta \ll 1$) is the region of rectangular pulses, for which V_{ptp} is independent of T. With T still increasing, V_{ptp} starts rising slowly until (at $T \approx 10\tau_T$) 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 τ_T and τ_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 $\tau_T = 2.4$ sec, and $\tau_e = 1.8$ msec. F_0 is 1 mW/cm^2, and the measured value k 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 $\tau_e \ll T_1 = T_2 = 54$ msec $\ll \tau_T$, and the response is seen to be close to rectangular. On Fig. 2(b) the chopping frequency is 59 Hz, and $T_1 = T_2 = 8.4$ msec. The exponential rise and decay of the response last for more than half the pulse duration. However, because $T_1 \nless t_p^+$, V_{ptp} is still the same as in the foregoing case. From the picture, one can estimate that V_{ptp} will remain frequency independent up to ~70 Hz. On Fig. 2(c) the chopping frequency is 380 Hz. $T_1 = T_2 = 1.3$ msec is slightly smaller than τ_e, and the response is already close to triangular. V_{ptp} 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 $R_L = 10^7\ \Omega$, so that $\tau_e = 0.32$ msec, and the condition $\tau_e \ll T_1, T_2$ is satisfied. V_{ptp} is seen to be independent of T_1/T_2 and equal 1.5 mV, as required by the formula $V_{ptp} = k\tau_e$. The traces of the middle column (traces e–h, 5 mV/cm vertical scale) were obtained with $R_L = 10^8\ \Omega$, $\tau_e = 3.2$ msec. On the two upper traces T_1 exceeds τ_e and even t_p^+; V_{ptp} 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 τ_e, but smaller than t_p^+. However, due to the flatness of the top of the response, V_{ptp} 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 i–l, vertical amplification 50 mV/cm) were obtained with $R_L = 10^{10}\ \Omega$ 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^+ of the rise increases, the slope k^- of the fall decreases (in absolute value), and so does V_{ptp}. The dependence of V_{ptp}, k^+, and k^- 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 τ_e. The switching of this time constant to τ_T when τ_e exceeds τ_T is shown in Fig. 6. The two traces here represent the pyroelectric responses in a SBN sample (B-1) of $C = 120$ pF and $\tau_T = 0.7$ 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 τ_e

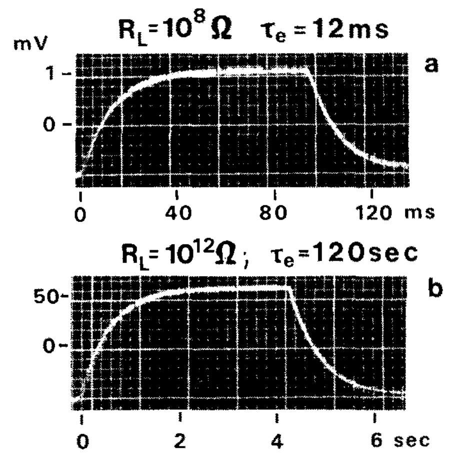

FIG. 6. Steady-state voltage response to rectangular radiation pulses in a SBN sample in case (a) $\tau_e \ll \tau_T$ and (b) $\tau_T \ll \tau_e$. SBN sample B-1: $F_0 = 0.5$ mW/cm^2; $\tau_T = 0.7$ sec; $C = 120$ pF.

$= 12$ msec $\ll \tau_T$, and the time constants of rise and fall are seen to equal τ_e. Trace b (1 sec/cm, 50 mV/cm) is for $R_L = 10^{12}\ \Omega$, so that $\tau_e = 120$ sec $\gg \tau_T$, and here both time constants equal τ_T. The initial slopes of rise and fall are 0.16 V/sec, independent of R_L. It is also seen, that while in the first case $V_{\text{ptp}} = 2$ mV $= k\tau_e$, in the second case $V_{\text{ptp}} = 110$ mV $= k\tau_T$.

VI. DISCUSSION AND CONCLUSIONS

The temperature rise of the sample has to be small during each single pulse, as well as during the whole train of pulses, so that ξ, τ_e, and τ_T remain constant and equal to their values in the step-signal case. Under these conditions, we have derived the pyroelectric voltage response to rectangular pulses from the response to step signals and have shown that the experimentally observed response in TGS and SBN samples follows the derived expression, though the uniformity of the absorption of radiation throughout the pyroelectric sample can be questioned.[6] The measured parameters of the response, such as rise and fall times, V_{ptp}, k^+, k^-, and their dependence on the parameters τ_e and τ_T of the sample, as well as on the parameters T_1/T_2 and frequency of the signal, are shown to be in good agreement with the analysis.

Similar to the step-signal response, the response to rectangular pulses is symmetric with respect to τ_e and τ_T. We have shown experimentally (on the rise and fall times and peak-to-peak values of the rectangular response, Fig. 6) that if, e.g., τ_e plays an important role in a certain parameter when $\tau_e \ll \tau_T$, then for $\tau_T \ll \tau_e$ the same role is played by τ_T. The response is also antisymmetric with respect to T_1 and T_2, so that interchanging the times of irradiation and darkening makes it flip around the time axis, leaving shape and peak-to-peak value unaltered. The values of T_1 and T_2, together with τ_e and τ_T, determine the shape of the response and the particular conditions under which it is rectangular or triangular.

The initial slope k of the response to a step signal is shown to be an important parameter of the response to pulsed excitation, too, especially in determining the slopes of rise and fall and the peak-to-peak values.

---

*Levy Eshkol Foundation Grantee of the Israel National Council for Research and Development.

[1] A. G. Chynoweth, J. Appl. Phys. 27, 78 (1956).
[2] J. Cooper, J. Sci. Instr. 39, 467 (1962).
[3] D. J. White, Naval Ordnance Laboratory Corona Reports No. 8170, 1963 (unpublished); Report No. 601, 1964 (unpublished); D. J. White and H. H. Wieder, J. Appl. Phys. 34, 2487 (1963).
[4] NASA Report No. NASA CR-44, 1964 (unpublished).
[5] A. Hadni, *Essentials of Modern Physics Applied to the Study of the Infrared* (Pergamon, New York, 1967); A. Hadni, R. Thomas, and J. Perrin, J. Appl. Phys. 40, 2740 (1969).
[6] M. Simhony and A. Shaulov, J. Appl. Phys. 42, 3741 (1971).
[7] This equality is obtained by integrating Eq. (2) of Ref. 6 during one cycle, which leads to the result $\int_t^{t+T} V\,dt = 0$.
[8] TGS crystals were laboratory grown; see V. Konstantinova, I. M. Silvestrova, and K. S. Aleksandrov, Sov. Phys. Crystallogr. 4, 63 (1960).
[9] SBN crystal courtesy of Dr. A. M. Glass of the Bell Telephone Laboratories, Murray Hill, N. J. (The Sr/Ba composition is 0.6/0.4, and the Curie point is ~90 °C.)

Structure and Composition of Sputtered Tantalum Thin Films on Silicon Studied by Nuclear and X-Ray Analysis*

M. Croset and G. Velasco†
Laboratoire Central de Recherches, Thomson-CSF, Domaine de Corbeville par Orsay 91, *France*
(Received 1 October 1971)

Microanalysis by direct observation of nuclear reactions and by backscattering of He_4^+ 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.[1,2] 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,[3-5] bcc,[5-11] tetragonal,[11-15] or amorphous.[4,5] 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 possibility of two crystalline forms: bcc (α-Ta) and tetragonal (β-Ta); the β system is associated with a higher resistivity and a TCR near zero; its occurence could be related to the purity of the deposit, the temperature, or the rate of deposition. Read and Altman[12] did not find any marked difference between the impurity content of β and α structures. Sosniak *et al.*[7] found that formation of β-Ta does not depend on the gas background in argon glow discharge. Gerstengerg and Calbick,[6] Krikorian and Sneed,[9] McLean,[10] and Baker[15] observed that, for defined sputtering conditions that normally produce the β form, the increase of oxygen or nitrogen concentration in the discharge atmosphere always produces the α phase. Later on, Westwood[13] and Westwood and Livermore[14] maintained that an increase of oxygen partial pressure in the discharge is associated with the forma-