Pyroelectric voltage response to short infrared laser pulses in triglycine sulphate and strontiumbarium niobate

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Analysis is given of the voltage response in a pyroelectric sample to ir signals much shorter than the electronic and thermal time constants of the sample. The equation of the response is derived, and the dependence of the various parameters of the response on the frequency and energy of the pulses as well as on the parameters of the sample and on material constants is compared with experiment in triglycine sulphate and strontium-barium niobate. The peak value of the response is found to be independent of the load resistance as well as of the pulse frequency, and proportional to the energy of the signals, provided that the mean temperature of the sample is far enough from the Curie point.

I. INTRODUCTION

The use of pyroelectric (PE) detectors[1,2] for the measurement of Q-switched ir laser pulses was reported by several workers.[3–6] Though some attention has been paid to the shape of the PE response in this case,[7,8] an adequate analysis has not been given yet.

Recently we have analyzed the PE voltage response to step ir signals.[9] An analysis is presented now of the PE voltage response to short ir laser pulses, "short" meaning that the pulse width is much smaller than the thermal and electronic time constants of the sample. The derived expressions for the peak value and other parameters of the response and their dependence on the frequency and energy of the pulses as well as on the parameters of the sample and its material constants are compared with experiment in triglycine sulphate[10] (TGS) and strontium-barium niobate[11] (SBN).

II. EXPERIMENTAL TECHNIQUE

The samples[9] were thin (10—40-μm) slices of TGS or SBN with evaporated InSb electrodes on their major faces perpendicular to the polar axis, mounted freely on 8-μm metal wire leads *in vacuo* in transistor cases with CdS windows.

The ir source was a model 42 Coherent Radiation Laboratories CO_2 laser (wavelength 10.6 μm) with a Q switch yielding ~1-μsec signals of repetition frequency 7.2 kHz, or with a pulser providing 100-μsec-wide signals of frequencies varying from 1 to 10^3 Hz. The energy of the radiation signals was attenuated with a set of calibrated optical filters and monitored by means of a beam splitter and power meter.

The measuring circuit consisted of a set of interchangeable load resistors (10^4—10^{11} Ω) connected parallel to the sample and to a simple FET source-follower circuit.[9] The PE response voltages were measured on Tektronix 535 or RM 564 oscilloscopes.

III. THEORETICAL ANALYSIS

Consider a PE sample with thermal and electronic time constants[9] $\tau_T = C_T/G_T$ and $\tau_e = RC$, C_T being the thermal capacitance of the sample, G_T its thermal conductivity to ambient, and R and C the resistance and capacitance of sample and circuitry at preamplifier input. The sample temperature is far enough from the Curie point so that all these values remain constant under small changes of temperature and voltage. Let a train of short ir signals of duration $\mathscr{T}_1 \ll \tau_T$ and τ_e and intervals $\mathscr{T} \gg \mathscr{T}_1$ be absorbed uniformly in the sample. If $\mathscr{T}$ does not exceed τ_T, the mean sample temperature $\bar{T}$ may be rising, but after a certain number of pulses a steady-state is achieved in which $\bar{T}$ remains constant. Owing to absorption of energy E_a from every signal, at the beginning of each period the sample temperature will rise suddenly by $\Delta T_0 = E_a/C_T$ above the level at the ends of the periods. If ΔT is the temperature increment above ambient, then during each interval $0 \le t < \mathscr{T}$ the cooling rate of the sample is $d(\Delta T)/dt = -\Delta T/\tau_T$. With the boundary condition $\Delta T(0) = \Delta T(\mathscr{T}) + \Delta T_0$, this yields

$$\Delta T = \Delta T_0 \exp(-t/\tau_T)/a_T, \tag{1}$$

where $a_T = 1 - \exp(-\mathscr{T}/\tau_T)$. Hence, during each interval $\mathscr{T}$ the temperature decays exponentially from $\Delta T(0) = \Delta T_0/a_T$ to $\Delta T(\mathscr{T}) = \Delta T_0(a_T^{-1} - 1)$ above ambient, with time constant τ_T.

The PE charge induced at $t=0$ is $Q_0 = \lambda AE/C_T$, where A is the electroded sample area, and $\lambda = dP_s/dT$ is the PE coefficient, with P_s the spontaneous polarization. The charge Q_0 raises the response at $t=0$ from the level $V(\mathscr{T})$ at the ends of the periods by V_0,

$$V_0 = \xi E_a C_s/AC. \tag{2}$$

Here C_s is the capacitance of the sample and $\xi \equiv \lambda/\epsilon c$ is a material constant,[9] ϵ being the permittivity, and c the

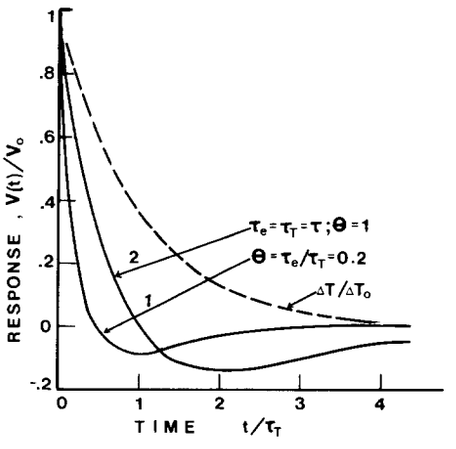

FIG. 1. Calculated time dependence of PE voltage response for two values of θ (normalized).

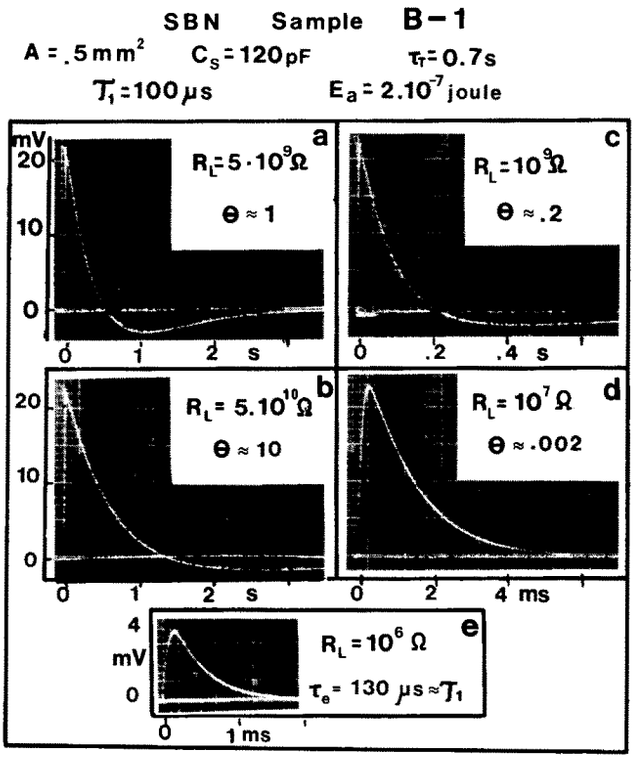

FIG. 2. Oscillograms of PE voltage response to single ir laser pulses in SBN sample with different load resistors.

volumetric specific heat of the material. V_0 is seen to be proportional to energy absorbed per unit area per pulse and to be independent of R. It also does not depend on $\mathscr{T}$, except through the dependence on temperature.

The disappearance of charge after the pulse is caused by flow through R and by change of P_s due to cooling. The PE voltage $V(t)$ therefore follows the equation

$$C\frac{dV}{dt}+\frac{V}{R}=\lambda A\frac{d(\Delta T)}{dt}. \tag{3}$$

Together with Eq. (1) and conditions $V(0)=V(\mathscr{T})+V_0$, $0\le t<\mathscr{T}$, this yields, for $\theta\equiv\tau_e/\tau_T\neq 1$,

$$V(t)=\frac{V_0}{\tau_T-\tau_e}\left(\frac{\tau_T}{a_e}e^{-t/\tau_e}-\frac{\tau_e}{a_T}e^{-t/\tau_T}\right), \tag{4}$$

where $a_e=1-\exp(-\mathscr{T}/\tau_e)$. $V(t)$ is symmetrical with respect to τ_e and τ_T. It decays from V_0 with an initial slope k,

$$k=\frac{-V_0}{\tau_T-\tau_e}\left(\frac{1}{\theta a_e}-\frac{\theta}{a_T}\right), \tag{5}$$

crosses the time axis at $t_1=\gamma\tau_e\ln(a_T/a_e\theta)$, where $\gamma=(1-\theta)^{-1}$, and reaches a minimum value V_m (if $\mathscr{T}$ is long enough),

$$V_m=-(V_0/\theta a_e)(\theta^{\theta}a_e/a_T)^{\gamma}, \tag{6}$$

at a time $t_m=\gamma\tau_e\ln(a_T/\theta^{\theta}a_e)$, thereafter returning slowly to the time axis. When τ_e and τ_T differ appreciably (θ far from unity), the decay of the response and the tail of its return from V_m to the time axis are exponentials whose time constants are, respectively, the smaller and the larger one from among τ_e and τ_T. This behavior is shown in Fig. 1 by curve 1, calculated for a single signal and $\theta=0.2$.

In case $\mathscr{T}\gg\tau_T$, τ_e, i.e., low signal repetition frequency f or single signal, $a_e=a_T=1$, so that

$$k=-V_0(\tau_T^{-1}+\tau_e^{-1}), \tag{5'}$$

$$V_m=-V_0\theta^{2\gamma-1}, \tag{6'}$$

and $t_m=2\gamma\tau_e(-\ln\theta)$.

For $\theta=1$, $\tau_e=\tau_T=\tau$, and $a_e=a_T=a=1-\exp(-\mathscr{T}/\tau)$, one obtains

$$V(t)=\frac{V_0}{a}\left(1-\frac{t}{\tau}+\frac{\mathscr{T}}{\tau}(1-a^{-1})\right)e^{-t/\tau}, \tag{4'}$$

$$k=-V_0\frac{2+(\mathscr{T}/\tau)(1-a^{-1})}{a\tau}, \tag{5''}$$

$$t_1=\tau+\mathscr{T}(1-a^{-1}),\quad t_m=t_1+\tau,$$

$$\text{and } V_m=(V_0/a)\exp[-2-(\mathscr{T}/\tau)(1-a^{-1})]. \tag{6''}$$

In the single-pulse or low-f case, $\mathscr{T}\gg\tau$, one obtains $k=-2V_0/\tau$, $t_m=2t_1=2\tau$, and $V_m=-V_0e^{-2}\approx-0.14V_0$. This case is represented by curve 2 of Fig. 1.

The "undershoot" value $|V_m|/V_0$ in the single-pulse or low-f case increases with θ from $|V_m|/V_0=\theta$ for $\theta\ll 1$ to the maximum of 14% for $\theta=1$, causing the peak-to-peak value V_{ptp} of the response to increase from V_0 to $1.14V_0$. In the intermediate frequencies, $\tau_e<\mathscr{T}<\tau_T$ (meaning also $\theta\ll 1$), $a_e\approx 1$, and $a_T\approx\mathscr{T}/\tau_T$, so that $V(0)=V_0(1-\tau_e/\mathscr{T})$ is smaller than V_0. However, if V_m is reached (i.e., $\mathscr{T}>t_m$), then $V_m=-V_0\tau_e/\mathscr{T}$ so that

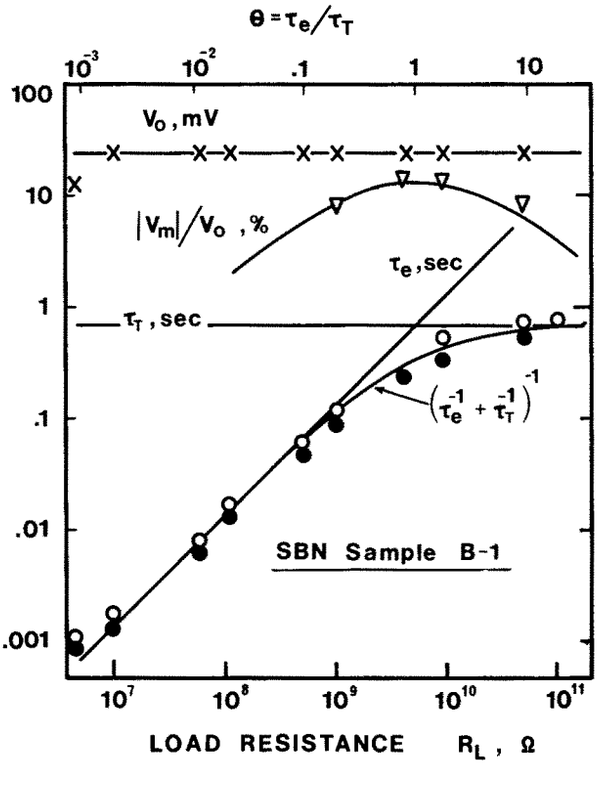

FIG. 3. Parameters of PE voltage response to single ir laser pulses in SBN sample as functions of R_L and θ.

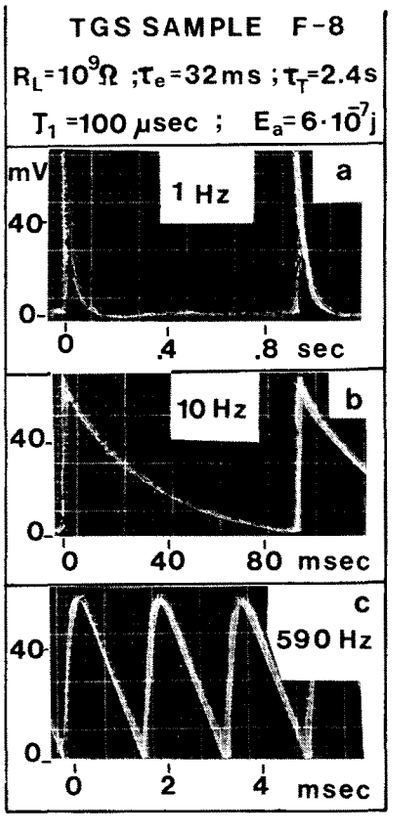

FIG. 4. Oscillograms of steady-state PE voltage response in TGS sample to a train of ir laser pulses of different frequencies. The 0-mV line is the $V(\mathscr{T})$ level.

$V_{ptp}=V(0)-V_m=V_0$. If V_m is not reached, i.e., at any frequency for which $\mathscr{T}<t_m$, $V_{ptp}=V(0)-V(\mathscr{T})\equiv V_0$.

At high frequency, $\mathscr{T}<\tau_T$ and τ_e, $a_T=\mathscr{T}/\tau_T-\frac{1}{2}(\mathscr{T}/\tau_T)^2$, $a_e=\mathscr{T}/\tau_e-\frac{1}{2}(\mathscr{T}/\tau_e)^2$, and $V(t)=V_0(\frac{1}{2}-t/\mathscr{T})$, i.e., the response is sawtooth shaped with $V(0)=-V(\mathscr{T})=\frac{1}{2}V_0$, $V_{ptp}=V_0$, and $t_1=\frac{1}{2}\mathscr{T}$. Here, as in all other cases, the areas enveloped by the response above and below the time axis are equal owing to charge conservation.

IV. EXPERIMENTAL RESULTS

Detailed studies of the PE voltage response to short pulses have been carried out at room temperature on 6 TGS samples and 3 samples of SBN. Typical responses to single CO_2 laser pulses (duration $\mathscr{T}_1=100$ μsec are shown by the CRO traces of Fig. 2(a)–2(d), obtained in a SBN sample B-1 with load resistors varying from 10^7 to 5×10^{10} Ω. The "short pulse" condition $\mathscr{T}_1\ll\tau_e$, τ_T holds, and it is seen that all responses rise abruptly to the same peak value. Trace a was obtained with $R_L=5\times10^9$ Ω to make θ close to unity. The steep rise of the response is followed by a decay with ~14% undershoot and a return to the time axis, as found by calculation (Fig. 1, curve 2). Trace b was obtained with $R_L=5\times10^{10}$ Ω, so that $\tau_e=6.5$ sec and $\theta\approx10$. The decay of the response is now nearly exponential with a fall time of 0.6 sec, close to τ_T. The undershoot is ~8%, and the return to the time axis (not completed on the picture) will proceed with a time constant close to τ_e. Trace c is with $R_L=10^9$ Ω, $\tau_e=130$ msec, and $\theta\approx0.2$. The decay is nearly exponential with a fall time of ~100 msec, close to τ_e. The undershoot is ~9%, and the return to the time axis will continue with a time constant close to τ_T. Trace d, with $R_L=10^7$ Ω, $\tau_e=1.3$ msec, and $\theta\approx0.002$, has an exponential decay with a 1.3-msec time constant equal to τ_e; no undershoot can be detected on this scale (its calculated value is 0.2%). In case e, $R_L=10^6$ Ω, $\tau_e=130$ μsec $\approx\mathscr{T}_1$, the pulse cannot be considered short, and our analysis is not applicable. The peak value of the response is seen to be smaller by a factor of 7 and will now decrease proportionally to R_L.

The parameters of the PE response in sample B-1 are plotted against R_L and θ in Fig. 3. The V_0 line was calculated from Eq. (2) using $C=130$ pF, $A=0.5$ mm^2, $E_a=2\times10^{-7}$ J, and $\xi=630$ cm^2/C.[9] Crosses show the measured values of V_0; they are independent of R_L over the whole range as long as τ_e, $\tau_T\gg\mathscr{T}_1$. The undershoot percent curve $|V_m|/V_0$ was calculated using Eq. (6). It is symmetric with respect to τ_e, τ_T and well matched by the experimental points (triangles). The $(\tau_e^{-1}+\tau_T^{-1})^{-1}$ curve was calculated using $\tau_T=0.7$ sec, and $\tau_e=R_L\times130$ pF.[9] The empty circles represent the measured fall times t_f of the response, while the full circles represent the ratios of the measured values V_0/k [see Eq. (5′)]. Both kinds of experimental points fit well the calculated curve and show the switching of t_f from τ_e to τ_T when $\theta\gg1$, i.e., t_f equals the smaller one from among τ_e and τ_T.

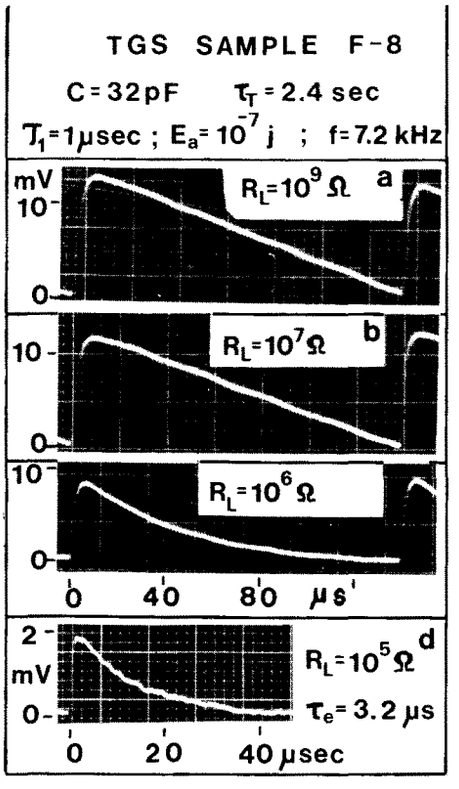

FIG. 5. Oscillograms of steady-state PE voltage response to a train of Q-switched ir laser pulses in TGS sample with different load resistors. The 0-mV line is the $V(\mathscr{T})$ level.

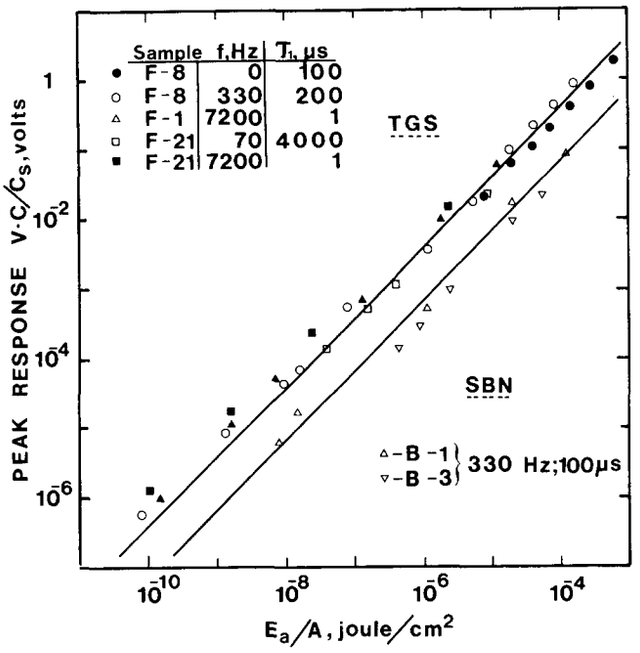

FIG. 6. Peak value of response vs ir signal energy (normalized). Lines are calculated from Eq. (2), for $C_s = C$ using $\xi = 4600$ cm²/C for TGS and 630 cm²/C for SBN.

The frequency dependence of the steady-state PE response to a train of short laser signals is represented by the oscillograms of Fig. 4. They were obtained in a TGS sample (F-8) with $R_L = 10^9$ Ω to make $\tau_e = 32$ msec $\ll \tau_T$; $\mathscr{T}_1 = 100$ μsec, so that $\mathscr{T}_1 \ll \tau_e$, τ_T. Trace a was obtained with $f = 1$ Hz, and it resembles the single-signal response. The 10-Hz frequency of trace b is intermediate, $\tau_e < \mathscr{T} < \tau_T$, whereas trace c (590 Hz) represents the high-frequency case with the sawtooth-shaped response. V_{ptp} is seen to be frequency independent over a four-orders-of-magnitude range.

The dependence of the high-frequency response on R_L is shown by the oscillograms of Fig. 5, obtained in sample F-8 with Q-switched signals of duration $\mathscr{T}_1 \approx 1$ μsec and $f = 7.2$ kHz. Traces a and b were obtained with $R_L = 10^9$ and 10^7 Ω, respectively. They are identical (small variations are due to the instability of the laser power), and identical responses were obtained also with $R_L = 10^{10}$ Ω. With $R_L = 10^6$ Ω (trace c) V_{ptp} is slightly decreased (9 mV instead of 12 mV), though the decay of the response changed to exponential ($\tau_e < \mathscr{T}$). With $R_L = 10^5$ Ω (trace d), $\tau_e = 3.2$ μsec, which is close to $\mathscr{T}_1$, and the short-signal approximation does not hold. V_{ptp} is smaller by a factor of 7 and continues to decrease with decreasing R_L.

The dependence of V_0 on the absorbed pulse energy is shown in Fig. 6. The lines here are calculated from Eq. (2), using[9] $\xi = 4600$ cm²/C for TGS (upper line) and 630 cm²/C for SBN. The experimental points were obtained in a number of samples with signals 1–4000 μsec wide, repetition frequencies 0 (single pulse), 330, and 7200 Hz, and absorbed energies varying from 10^{-10} to 10^{-4} J/cm² per pulse. E_a was calculated from the power-meter readings and the $\mathscr{T}_1/\mathscr{T}$ ratio, assuming a Gaussian distribution of energy around the axis of the laser beam and reducing ~40% for losses in the window and reflection from the front electrode. The points are seen to fit the calculated lines within a factor of 2 over the whole range of energies, while the linearity of the dependence for most samples is within 10%.

V. DISCUSSION AND CONCLUSIONS

The short-signal condition $\mathscr{T}_1 \ll \tau_T$, τ_e can be satisfied for a wide range of signal widths, mostly by adjusting R_L so that $\tau_e \gtrsim 10\mathscr{T}_1$. While τ_T is 30 msec to 3 sec, depending on construction, and remains a sample constant, τ_e can easily be changed by many orders of magnitude; e.g., in a sample with $C_s = 10$ pF and $R_s = 10^{13}$ Ω, appropriate load resistors can make τ_e vary between 10 nsec and 10 sec. Under all circumstances $R_L \ll R_s$, so that $R = R_L = \text{const}$.

The analysis assumes that ϵ, λ, and c are constant *during* the pulses, which holds if ΔT is small and $\bar{T}$ is far from the Curie point. For single signals or low f this was obeyed with pulse energies up to 10^{-3} J/cm². (For example, in TGS sample F-8, then $\Delta T = 0.2$ °C.) The rise in $\bar{T}$ was limited to ~10 °C so that in most TGS samples E_a/A did not exceed 10^{-4} J/cm² with $f = 330$ Hz, or 4×10^{-6} J/cm² with $f = 7.2$ kHz. In SBN the energies could be increased by a factor of 2.

The expression for the PE response was derived assuming uniform absorption of ir radiation throughout the sample.[1] Experimentally observed responses in TGS and SBN follow this expression, though the uniformity of absorption can be questioned.[9]

The independence of V_{ptp} on R_L starts from R_L such that $\tau_e \gtrsim 10\mathscr{T}_1$ and extends over many orders of magnitude. Only in the low-f case when R_L is so large that τ_e approaches τ_T, V_{ptp} will rise by up to 14%. The increase of R_L affects the shape of the response (undershoot, change of decay from exponential to linear, etc.), but has no effect on the peak value.

Similar changes are observed when f is increased. V_{ptp} remains constant as long as $1/f = \mathscr{T} \gg \mathscr{T}_1$, i.e., in an f range starting from 0 (single-signal) up to 10 kHz with 1-μsec signals, and to even higher f with correspondingly shorter signals.

The linearity of V_s vs E_0/A suggests that the PE detection of short signals should be characterized by a minimum detectable energy rather than power; e.g., if the electronics accepts 50-nV responses from TGS samples, which means a minimum detectable energy of $E_a/A \approx V_0/\xi = 10^{-11}$ J/cm², then, for 1-μsec signals, the minimum detectable power is 10^{-5} W/cm², while for 1-msec signals it is 10^{-8} W/cm².

Here, and throughout, we disregarded the dependence of ξ on the frequency ω (reciprocal rise time of the signals). The dependence of ϵ, λ, and c on ω in the range under consideration is rather weak,[12] and the changes it may cause in the parameters of the response are within the experimental error.

It should be pointed out that the response to short signals [Eq. (4)] can be derived by differentiation of the response to a step signal.[9] The independence of V_0 on R is then a consequence of the fact that the initial slope

of the step-signal response does not depend on R. Also, it was shown by Putley[1] and by Hadni *et al.*[7] that for high-frequency periodic radiation, the response is independent of load resistance and proportional to power/frequency, which is the same as energy per cycle or per pulse.

ACKNOWLEDGMENT

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Analysis of triple dislocations in ordered phases of the type Ni_8X

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Superdislocations comprising a group of three $(1/2)a\langle 110\rangle$ dislocations lying in a close-packed plane have been observed in ordered face-centered-tetragonal Ni_8Ta and Ni_8Nb alloys. The antiphase boundary (APB) energies of the areas between the dislocations have been evaluated for Ni_8Ta in terms of the first- and higher-order nearest-neighbor atom pairs associated with the introduction of the dislocations in the ordered material. Values for the pair interaction have been calculated by employing a polar model for the ordering and a long-range oscillatory atomic pair interaction energy. The results indicate that higher than first nearest-neighbor interactions account for approximately 40% of the total ordering energy. The critical temperature of Ni_8Ta has been estimated on the basis of Cowley's ordering theory and the calculated values of the pair interaction energies. The theoretical value for the critical temperature is within 12% of the value that was determined by experiment.

INTRODUCTION

A new type of long-range order (LRO) has been reported to occur in Ni-rich Ni-Nb and Ni-Ta alloys[1,2] near the stoichiometric Ni_8X composition. From the face-centered-tetragonal (fct) Ni_8X unit cell of the ordered structure shown in Fig. 1, it can be seen that the X atoms are at relatively large distances from each other, indicating that long-range atomic interactions assume an important role in this type of order. Triple dislocations have been observed in the ordered Ni_8X structure[3]; their occurrence in both symmetric and asymmetric configurations is consistent with the atomic periodicity of the close-packed layers and with the anisotropy of the ordered structure. The characteristics of these dislocations can be interpreted on the basis of Fig. 2 which represents the (111) close-packed plane of the ordered structure. The configuration of these triple dislocations has been used in the present paper as a basis for evaluating the antiphase boundary (APB) energy from which the atomic pair interaction energy terms for the Ni_8Ta alloy have been calculated. The paper also presents an adaptation of Cowley's ordering theory[4] which is used to calculate a theoretical value of the ordering temperature for the stoichiometric Ni_8Ta alloy.

EXPERIMENTAL PROCEDURES

The Ni—11.1-at.% Ta alloy was prepared by levitation melting and chill casting in a purified He atmosphere. Samples of the alloy were swaged into 2.3-mm-diam rods. A vertical quenching furnace was employed for the solution treatment temperatures above 1300 °C and permitted the rapid quenching of the rods to room temperature by immersion in a 10-wt.% NaOH solution. The specimens were then aged in a molten salt bath or vacuo at temperatures in the range of 400—650 °C for times up to 4000 h. The heat-treated rods were cut into wafers 0.5 mm thick, from which thin foils were prepared for electron microscopy by means of an automatic electropolishing technique.[5]

Electrical resistivity measurements were used to monitor the degree of long-range order (LRO) of the Ni_8Ta rods during the aging treatments. Short chromel potential and current leads were resistance welded to 10-cm-long rod specimens after quenching from the solution treatment temperature. The specimen was placed in series with a 1-Ω standard resistor and connected to a Sorensen QRB power supply which provided a constant current of 0.2 A. The specimen was then immersed in liquid nitrogen, and the voltage drops across