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Theoretical Considerations on the Switching Transient in Ferroelectrics

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Calculations are made to find an analytical expression for the switching transient in different ferroelectrics. By studying this expression, it is possible to give a definition of the nucleus-domain interaction and to find a numerical relationship between this interaction and the shape of the switching pulse. An attempt is made to explain the power-law dependence of the switching time on applied field, which was found experimentally in $BaTiO_3$ (at high fields only), in thiourea, and in other ferroelectrics.

1. INTRODUCTION

IT is possible to study the reversal of the spontaneous polarization in a ferroelectric material by measuring the current i_s, which flows during switching as a function of time t. The shape of the i_s vs t curve (switching pulse) depends on the material and is a function of the applied field E.

It is difficult, however, to study the switching pulse, as this varies widely in shape under different conditions. For this reason, most papers[1–20] have dealt with just one characteristic of the switching pulse, its duration t_s, which is the time necessary to switch the ferroelectric (switching time).

In some papers,[15,18,19,21] however, the shape of the switching pulse has been studied in more detail. Peacock[19] and Wieder,[18] for instance, calculated the shape of the switching pulse under the assumption that there is sideways expansion of the domains during switching. They also made the approximation that the sideways wall velocity is so high as to make the size of the original nucleus negligible with respect to that of the final domain. This approximation appears to be valid for the two substances for which it was made ($BaTiO_3$ and colemanite) but leads to results which do not apply to all ferroelectrics. For instance, it follows from Peacock's and Wieder's calculations that the shape of the switching pulse is "symmetrical." This is not true with triglycine sulfate, the reason being in our opinion, that the sideways motion in triglycine sulfate is not very fast; the original size of the nucleus is therefore not negligible with respect to that of the final domain as a consequence of sideways motion.

In the course of this paper we shall take the original size of the nucleus into account and we shall show that the shape of the switching pulse in triglycine sulfate and other ferroelectrics can then be explained.

A paper by Fatuzzo and Merz[15] considered the general case in which the sideways motion (called "nucleus-domain interaction" in the paper) can be both fast and slow. It was found that a slow sideways motion (weak domain-nucleus interaction) yields a very "asymmetrical" pulse, whereas a fast sideways motion yields a "symmetrical" pulse. The paper of Fatuzzo and Marz, however, did not provide: (1) a suitable analytical expression which describes the switching pulse; (2) a more precise definition of the nucleus-domain interaction; (3) the relationship between this interaction and the symmetry of the switching pulse at low fields; (4) an explanation for the power law dependence of the switching time t_s on field E found in $BaTiO_3$ at high fields,[22] in thiourea,[20] in $LiH_3(SeO_3)_3$,[14] and in tetramethylammonium-trichloro-mercurate (TTM).[17] These points can only be explained by making more accurate calculations which consider the mechanism of switching in more detail. Before proceeding with these calculations, however, it is necessary to make definite assumptions about the switching mechanism.

We shall assume that the nuclei of domains are formed at random on the surface of the crystal according to a statistical process. The time t_n necessary to form *all* the nuclei (from the first to the last) is given by the exponential law[15]:

$$1/t_n = (1/t_0)\exp(-\alpha/E). \qquad (1)$$

When formed, each nucleus grows in the direction of the field in a time t_d, until it becomes a domain and reaches the opposite electrode. The domain formed in this way starts expanding sideways with velocity V. During this expansion, a new domain is formed from a new nucleus and also starts growing sideways (Fig. 1); then a third domain is formed and so on. After some time these domains are large enough to join each other, or to "coalesce" until, through successive nucleation, sideways growth and coalescence, the polarization is com-

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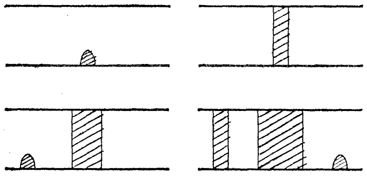

FIG. 1. Drawing demonstrating the growth of domains by forward and sideways motion.

pletely reversed in the whole sample (Fig. 1). We have not yet specified how this sideways motion takes place and shall now consider two possible mechanisms. The first is that the domain expands through a true sideways motion; the dipoles adjacent to the 180° domain walls switch one row at a time [Fig. 2(a)]. The alternative is that the edge of an existing domain is more favorable for the nucleation and growth of new domains than elsewhere.[23] Since this effect also has the appearance of a sideways motion [Fig. 2(b)] we shall refer to the first case as a "true" and to the second as an "apparent" sideways motion.

Throughout this paper we shall assume the above picture for the switching of ferroelectrics. We shall furthermore assume that the forward-domain wall-motion time t_d is much shorter than the nucleation time t_n. The switching current i_s is then at each instant proportional to the rate of nucleation.[15] In $BaTiO_3$ this assumption is valid for applied fields lower than 50 kV/cm[15] and in triglycine sulfate for fields lower than 5 kV/cm.

The paper is divided into three main parts: In Sec. 2 the basic equation which gives i_s as a function of time t is derived. In Sec. 3 the results are examined and the consequences of the basic equation discussed. In Sec. 4 some conclusions are drawn.

2. BASIC EQUATION

2.1 Area of One Domain as a Function of Time

The volume of a domain is given by σl, where σ is the area intersected by the domain with the electrode and l is the crystal thickness (Fig. 2). Since l is a constant, we can confine ourselves to the areas σ. We would like to calculate σ as a function of time, under the two different assumptions that the domains expand sideways by *true* motion or by *apparent* motion. In the case of apparent motion, the radius r of a domain (supposed to be circular) is assumed to increase with time according to the equation

$$r=r_c+(2r_c)R^*t,$$

where t is the time, r_c is the radius of the nucleus at its birth, and R^* is the probability per unit time that the radius of the domain increases by the amount $(2r_c)$. In the case of true motion, we assume that the radius r of the domain increases according to the equation[24]

$$r=r_c+\mu_t Et,$$

where μ_t is the sideways mobility and E the applied field. In this last equation, we have neglected the inertia of the wall, because no evidence for this inertia has been observed experimentally.[25–28] If T is the total electrode area, it can be shown that the fractional area $\sigma=\pi r_c^2/T$ is described in both the cases by the following equation:

$$\sigma=(v_0^2/T)[t_0+t]^2, \tag{2}$$

where

$$v_0^2=\pi\mu_T^2E^2, \quad t_0=r_c/\mu_t E \tag{3}$$

in the case of true sideways motion and

$$v_0^2=4\pi r_c^2R^{*2}, \quad t_0=1/2R^* \tag{4}$$

in the case of apparent sideways motion. In Eq. (2), the quantity v_0 represents the sideways wall velocity. If we subtract from σ the (fractional) area σ_c of a nucleus at its birth, we obtain the area σ_n which is the amount the domain has expanded through sideways motion (Fig. 3). Since

$$\sigma_c=\pi r_c^2/T,$$

we have

$$\sigma_n=\sigma-\sigma_c=(v_0^2/T)[t_0+t]^2-\pi r_c^2/T. \tag{5}$$

2.2 Switched Area as a Function of Time

In this section the area covered by the switched domains will be calculated as a function of time t, with the two following assumptions: (a) The possibility of over-running of nucleation sites by the sideways growth of domains is neglected. (b) The coalescence of the domains due to their sideways expansions is neglected. The area calculated under these restrictive conditions will be called A.

Although the above two assumptions are not realistic, *the calculation of the area A is necessary for the subsequent and more accurate calculation.* We shall then proceed to calculate the area covered by the switched domains, without the two restrictive conditions mentioned above; this area will be called θ. A theorem due to Avrami[29] allows the calculation of θ, once A is known. It will be seen later that A and θ almost coincide at the beginning

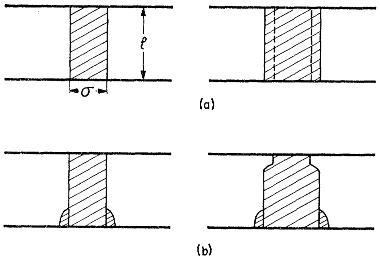

FIG. 2. (a) Drawing demonstrating the sideways expansion of a domain by *true* motion. (b) Drawing demonstrating the sideways expansion of a domain by *apparent* motion.

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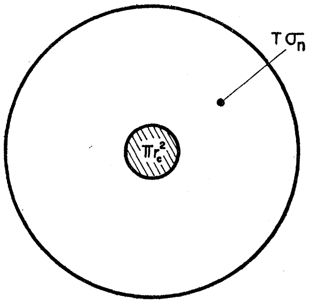

FIG. 3. Drawing demonstrating the original size πr_c^2 of the nucleus (shaded area) and the amount σ_n the domain has grown by sideways motion.

of the switching. However, A tends to infinity, while θ tends to the electrode area when $t \to \infty$. If we refer to unit area, we have

$$\lim_{t\to\infty} \theta = 1.$$

Let N_0 be the total number of nucleation sites (i.e., the total number of nuclei that can be formed) and $N(t)$ the actual number of domains formed at the time t. We assume that the nuclei are formed according to a statistical process[15] in a random way, with a probability R per unit time. We can then write

$$dN/(N_0-N) = Rdt,$$

and hence by integrating this equation

$$N = N_0(1-e^{-Rt}). \qquad (6)$$

The area A can now be easily calculated:

$$A = \int_0^t \left(\frac{dN}{dt}\right)_s (\sigma_n)_{t-s} ds + \frac{\pi r_c^2}{T} N(t). \qquad (7)$$

Here, the integral represents the area A covered by the *growth* of domains, while the second term in the sum represents the area covered by the N new nuclei at their birth. In the integral, the term $(dN/dt)_s$ is the rate of nucleation at the time s and can be calculated from Eq. (6). The term $(\sigma_n)_{t-s}$ represents the area σ_n at the time $(t-s)$ and can be computed from Eq. (5). Evaluating the integral and rearranging the terms, we obtain

$$A = \frac{2N_0 v_0^2}{TR^2}\Big[1 - R(t+t_0) + \tfrac{1}{2}R^2(t_0+t)^2 - e^{-Rt}(1 - Rt_0 + \tfrac{1}{2}R^2 t_0^2)\Big] - \frac{\pi r_c^2 N_0}{T}(1-e^{-Rt}) + \frac{\pi r_c^2}{T} N(t). \qquad (8)$$

The value of A given by Eq. (8) tends to infinity when $t \to \infty$, which is, of course, physically impossible. The reason for this is that the coalescence of domains and overrunning of nucleation sites has not been considered.

The area θ can be calculated from the area A, using the theorem demonstrated by Avrami.[29] This theorem, however, neglects the area covered by the nucleus at its birth and must therefore be slightly modified to meet our purposes. By making this modification, it is found (see Appendix, Eq. 30) that

$$\theta = 1 - \exp\left\{-\left[A + \frac{\pi r_c^2 N_0 Rt}{T} - \frac{\pi r_c^2}{T} N(t)\right]\right\}. \qquad (9)$$

Therefore, substituting Eq. (8) into this equation and rearranging terms, one obtains (see again Appendix)

$$\theta = 1 - \exp\{-2k^2[1 - (\tau + k^{-1}) + \tfrac{1}{2}(\tau + k^{-1})^2 - e^{-\tau}(1 - k^{-1}) - \tfrac{1}{2}k^{-2}(1-\tau)]\}, \qquad (10)$$

where

$$k = 1/Rt_0, \qquad (11)$$

$$\tau = Rt. \qquad (12)$$

Here, τ can be considered a kind of "dimensionless time."

The current i_s which flows during switching is then given by

$$i_s = 2P_s(d\theta/dt) = 2P_sR(d\theta/d\tau),$$

so that

$$i_s = 4P_sRk^2[-1 + (\tau + k^{-1}) + e^{-\tau}(1 - k^{-1}) + \tfrac{1}{2}k^{-2}] \times \exp\{-2k^2[1 - (\tau + k^{-1}) + \tfrac{1}{2}(\tau + k^{-1})^2 - e^{-\tau}(1 - k^{-1}) - \tfrac{1}{2}k^{-2}(1-\tau)]\}. \qquad (13)$$

It can be seen that Eq. (13) fully describes the shape of the switching pulse. From Eq. (13) one sees that, when $k \to 0$, the switching current i_s as a function of time is given by

$$i_s = 2P_sRe^{-\tau}.$$

This exponential dependence of the current i_s is exactly what is expected when the domain-nucleus interaction is zero. [Cf. Eq. (26) of reference 15.] When $k \to \infty$, Eq. (13) becomes

$$i_s = 4P_sRk^2[-1 + \tau + e^{-\tau}] \times \exp\{-2k^2[1 - \tau + \tfrac{1}{2}\tau^2 - e^{-\tau}]\}, \qquad (14)$$

which is the same as the equation found by Wieder[18] and Peacock[19] for the case when k is very large.

3. DISCUSSION

Starting from Eq. (13) we can draw a number of conclusions. The nucleus-domain interaction, the switching time, the shape of the switching pulse, and the dependence of the switching time t_s on applied field E will all be discussed separately.

3.1 Nucleus-Domain Interaction

From Eq. (13) it appears that the normalized shape of the switching pulse depends only on the parameter $k = 1/Rt_0$. From Eqs. (3) and (4), it can be shown that this parameter can be written

$$k = \mu_t E / Rr_c \qquad (15)$$

for the case of a true motion, and

$$k=2R^*/R \tag{16}$$

for the case of an apparent motion.

From Eq. (16) it appears that, in the case of apparent motion, $k/2$ is given by the probability R^* that the new domain is formed near an existing domain, divided by the probability R that it is formed somewhere else. It appears, therefore, that k can be identified with the "nucleus-domain interaction" of reference 15. From this it follows that the shape of the switching pulse depends just on the nucleus-domain interaction k as was proposed in reference 15. On the other hand, Eq. (13) is also valid for the case of true sideways motion, provided the dimensionless constant k is computed from Eq. (15) and not from Eq. (16). It follows that all the conclusions of reference 15, which were reached under the assumption of an apparent motion, must also be valid in the case of true motion.

As mentioned at the beginning of this section, the shape of the switching pulse depends only on the parameter k. When k is small ($\leqslant 1$), the shape of the pulse depends rather markedly on k. When k is large [Eq. (14)] it can be shown that the shape of the pulse is fairly independent of k. Since the pulse shapes observed experimentally in different ferroelectrics do not vary much as a function of the applied field E,[30] we may also conclude that k does not depend much on E. This conclusion, however, is true only for those materials in which k is *not* much greater than 1; if k is much greater than 1, this quantity can vary with field without producing changes in the switching pulse.

3.2 Switching Time

In this section we calculate the dependence of the switching time on the field E in two particular cases:

(a) $k \gg 1$;

(b) k is not much larger than 1, but it is constant with field. Although these are only two particular cases, in practice they cover the majority of the experimental cases.

3.2.1 $k \gg 1$

The assumption that $k \gg 1$ implies that the sideways motion is a predominant mechanism of switching. In order to calculate the dependence of t_s on E, we redefine the "switching time" t_s as the time necessary to reverse 95% of the polarization of the sample. The problem is to find how t_s varies with k. If $\tau_s = Rt_s$ is a "dimensionless switching time," we can write for the area θ at the time τ_s:

$$\theta(\tau_s)=0.95,$$

because 95% of the polarization is switched. By substituting this value into Eq. (10), we find

$$2k^2\left[1-(\tau_s+k^{-1})+\tfrac{1}{2}(\tau_s+k^{-1})^2 - [\exp(-\tau_s)](1-k^{-1})-\frac{1}{2k^2}(1-\tau_s)\right]\cong 3, \tag{17}$$

from which it follows that

$$\lim_{k\to\infty} \tau_s = 0. \tag{18}$$

When k is large enough, Eq. (17) can be written as

$$2k^2[1-\tau_s+\tfrac{1}{2}\tau_s^2-\exp(-\tau_s)]\cong 3. \tag{19}$$

Since $\tau_s \ll 1$ when k is sufficiently large [see Eq. (18)], we can write

$$\exp(-\tau_s)\cong 1-\tau_s+\tfrac{1}{2}\tau_s^2-\tfrac{1}{6}\tau_s^3,$$

and hence from Eq. (19) it follows that

$$\tau_s=(3/k)^{\frac{2}{3}}\cong 2k^{-\frac{2}{3}}. \tag{20}$$

We can now use Eq. (20), in which k is given by Eq. (15) in the case of true sideways motion and by Eq. (16) in the case of apparent sideways motion. It must be remembered that R, R^*, and r_c are field dependent and that their exact dependence on field must be known in order to calculate τ_s. We shall assume therefore[1,7,18,19] that

$$R \propto \exp(-\alpha'/E) \tag{21}$$

and

$$R^* \propto \exp(-\alpha''/E). \tag{22}$$

The dependence of r_c on E is related to the model assumed for the nucleation. If we assume a nucleus with circular base, and take the depolarizing energy into account,[7] we have

$$r_c \propto E^{-1}. \tag{23}$$

With other models for the nucleation we have a dependence of r_c on E of the type $r_c \propto E^{-m}$, where m is always a small number (1 or 2).

Taking the values of R, R^*, and r_c given by Eqs. (21), (22), and (23) into account, and substituting them into Eqs. (15) and (20), we obtain for the case of *true* sideways motion:

$$\tau_s \propto [E^2/\exp(-\alpha'/E)]^{-2/3} = E^{-4/3}\exp(2\alpha'/3E). \tag{24}$$

But $\tau_s = Rt_s$, so that (still in the case of *true* motion) we can write Eq. (24) as follows:

$$t_s = E^{-1.33}e^{5\alpha'/3E} = E^{-1.33}\exp(\alpha^*/E), \tag{25}$$

where $\alpha^* = 5\alpha'/3$. In the case of apparent sideways motion we have instead

$$t_s \propto \exp[(2\alpha''/3E)+(\alpha'/3E)] = \exp(\bar{\alpha}/E),$$

where

$$\bar{\alpha} = (2\alpha''/3)+(\alpha'/3).$$

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[30] We do not consider the change in shape due to the forward growth of domains, that was studied in reference 15.

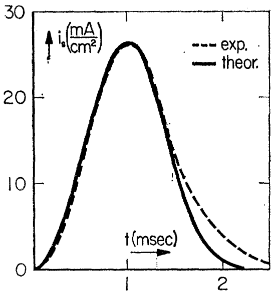

FIG. 4. Experimental and theoretical switching pulse in $BaTiO_3$.

Eq. (25) represents the cases of $BaTiO_3$ or thiourea: For low values of the field the exponential term is predominant, because it varies much more rapidly than $E^{-1.33}$. At high fields the exponential term saturates and the term $E^{-1.33}$ takes over. It is stressed that this equation could provide an explanation for the power-law dependence of t_s on E found by Stadler[22] in $BaTiO_3$ at high fields and by Goldsmith[20] in thiourea. Equation (25), however, is valid just in the case of *true* sideways motion; but all the experimental evidence, available at low fields, seems to indicate the presence of an apparent sideways motion. We could resolve this difficulty by assuming that the nature of the sideways motion changes from *apparent* to *true*, in going from low to high fields.

However, we do not have any real evidence of this change from apparent to real sideways motion. But this picture provides an explanation for the power law which in our opinion seems more likely than others.[31]

3.2.2. $k=$ *const*

In this case k is not much larger than 1; this means that the sideways motion is *not* the predominant mechanism of the switching. Furthermore, it has been assumed that k is almost constant with field. This rules out the possibility that the sideways motion is of the *true* type [see Eq. (15)]. In order to fulfill this assumption, we must have apparent sideways motion [see Eq. (16)] and the two coefficients α' and α'' [see Eqs. (21) and (22)] must be approximately equal. This is not unreasonable because both R^* and R are nucleation probabilities in the same crystal. If the ratio R^*/R is constant with field, it means that the field dependences of R^* and R are the same, and only the proportionality constants are different. We may assume that this case does occur, because in several ferroelectrics we do observe a switching pulse whose shape (and hence k) is field independent. Examples of such ferroelectrics are triglycine sulfate (TGS) and guanidine aluminum sulfate hexahydrate (GASH).

After having discussed the physical implications of our assumption, we may now proceed with the calculations. Since k is constant with field, we may now solve Eq. (17) for τ_s and we obtain

$$\tau_s = f(k) = \text{const},$$

where $f(k)$ is a function of k alone and hence is a constant, like k. Since $\tau_s = Rt_s$, we may write [see Eq. (21)]

$$t_s = \text{const}R = \text{const}e^{-\alpha'/E}.$$

This result explains the switching in TGS and GASH (at low fields) where the assumption (b) above is satisfied. In both TGS and GASH an exponential law for the switching time is found.

3.3 Shape of the Switching Pulse

We can now study the dependence of the shape of the switching pulse on k and we may compare the theoretical switching pulse given by Eq. (13) with the experimental cases.

When k is very large, Eq. (13) becomes identical with Eq. (14) which is the equation found by Wieder[18] and Peacock.[19] Wieder[18] has shown that this equation fits the experimental curve in the case of colemanite (see Fig. 9 of reference 18).

In Fig. 4 we have plotted both Eq. (13) with $k \gg 1$ and an experimental pulse of $BaTiO_3$. The agreement is fairly good. In both the cases of $BaTiO_3$ and colemanite the experiment and theory agree if we assume that $k \gg 1$, namely, that the sideways motion is the important mechanism in the switching.

At the other extreme, we have the case of GASH,[16] where experiment and theory agree if we take $k \cong 0$. The experimental points are compared with the theoretical curve in Fig. 5. In this compound almost no sideways motion of domain occurs; the switching takes place mainly through the forward growth of many small domains.

The two intermediate cases are TGS and TTM. In TGS, the experimental and theoretical curve agree if we take $k=1$ (see Fig. 6), and in tetramethylammonium trichloro mercurate (TTM), if we take $k=5$ (see Fig. 7). In both these cases, forward and sideways growth of domains are of comparable importance in the switching,

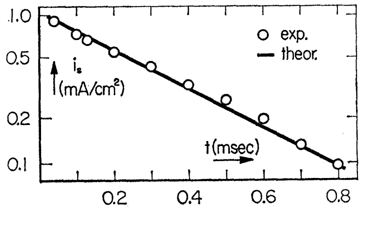

FIG. 5. Experimental and theoretical switching pulse in guanidine aluminum sulfate hexahydrate (GASH).

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[31] If the probability R^* to create a nucleation center in the vicinity of an existing domain were field dependent with an extra term, for instance $R^* \propto E^\lambda \exp(\alpha'/E)$, this extra term would also lead to a power law for the switching time. However, this extra term E^λ would be difficult to be explained for the following reasons: All the expressions for nucleation probability usually lead to a pure exponential.[1,7] Furthermore, if this term E^λ existed, it would also appear in the expression for the sideways velocity of domains (since $R^* \propto v_0$) and this is not found experimentally,[25,28] at least at low fields.

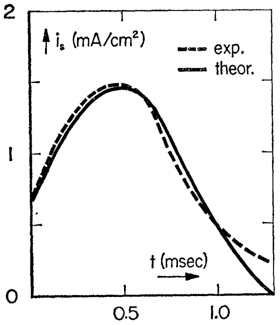

FIG. 6. Experimental and theoretical switching pulse in triglycine sulfate (TGS).

the sideways motion being more important in TTM than in TGS.

A qualitative explanation may reveal a better insight in the problem than the quantitative treatment made so far. If the switching occurs predominantly through sideways motion (e.g., $BaTiO_3$), the switching current is at any instant proportional to the domain perimeter l. Since $l=0$ at the beginning of the switching, the switching current i_s is equal to zero. It then increases, passes through a maximum and decreases again. If on the other hand, the switching occurs through nucleation and forward growth of domains (as in GASH), the switching current is proportional to the rate of nucleation and is a maximum at the beginning of the switching. In intermediate cases (like TGS or TTM), the switching current has a value different from zero at the beginning of the switching, then it increases, goes through a maximum and decreases again. The ratio of the current i_0 at the beginning of the switching to the maximum switching current $i_{\max}$ could be a good way to determine the relative importance of the sideways motion during the switching. Unfortunately, however, the value i_0 is not easily measurable because it is masked by the initial "capacitance peak," and therefore the determination of the "symmetry" of the pulse is a better way to determine the extent of the sideways motion.[15]

4. CONCLUSIONS

The calculations presented in this paper seem to answer some of the questions formulated in Sec. 1. Eq. (13) is an analytical expression for the switching current i_s as a function of time. From this expression it appears that the shape of the switching pulse depends only on a quantity k, which can be identified with the "nucleus-domain interaction" of reference (15). The analytical expression of Eq. (13) compares fairly well with the experimental low-field switching pulses in a number of ferroelectrics: colemanite[18] and $BaTiO_3$[19] (assuming $k\gg 1$), TTM (assuming $k=5$), TGS (assuming $k=1$), and GASH (assuming $k\cong 0$).

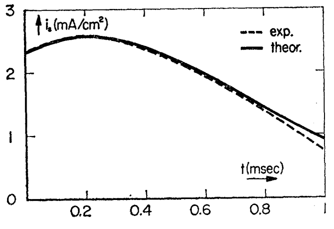

Fig. 7

FIG. 7. Experimental and theoretical switching pulse in tetramethylammonium trichloro mercurate (TTM).

The exponential law dependence of the switching time on field observed experimentally at low-field values is also explained by the present model.

An attempt has also been made to explain the power law dependence of t_s on E, found in many ferroelectrics. Eq. (25) might explain the power law found in $BaTiO_3$[22] and thiourea,[20] if we assume that the sideways motion changes from "apparent" to "true" when going from low to high fields. This, however, is still a rather speculative explanation and we have no real proof for it yet. If we disregard these difficulties we are led to an attractive model for the power-law dependence of t_s on E, namely,

(1) At very low fields, at which the switching is controlled by nucleation of new domains, we obtain an exponential field dependence of t_s on E.

(2) At intermediate fields, at which the switching is controlled by the forward growth of the domains, we have proportionality of $1/t_s$ to E.

(3) At very high fields, when switching is controlled by the forward growth of the domains, we have proportionality of $1/t_s$ to E.

Range 3 might occur at fields too high to be attainable (as in $BaTiO_3$ and thiourea[20]); range 2 might be hidden if ranges 1 and 3 are too close to each other (as in triglycine sulfate).

APPENDIX

In this Appendix we want to demonstrate the validity of Eqs. (9) and (10). Let us first adapt the theorem of Avrami to our case, where the nucleus size cannot be neglected. First of all we introduce the following definitions:

We divide the area $A(t)$ into two parts $A(t)=A^*(t)+A_n(t)$, where

$$A_n(t)=\sum\sigma_n(t)$$

is the fractional area covered by all the domains due to their sideways growth and

$$A^*(t)=\sum(\pi r_c^2/T)=(\pi/T)r_c^2N(t)$$

is the fractional area covered by all the nuclei. In both cases the sum is extended to all the domains formed at the time t. From the last two equations it follows that:

$$A=A_n+A^*=A_n+(\pi/T)r_c^2N(t). \tag{26}$$

Let us also divide the area into two parts: θ^* and θ_n, where θ^* corresponds to A^* and θ_n corresponds to A_n.

According to Avrami's notation, for each domain we call[29] "extended volume" the volume which the domain would have if it had extended without meeting any other domain (Fig. 8); the "nonoverlapped volume"

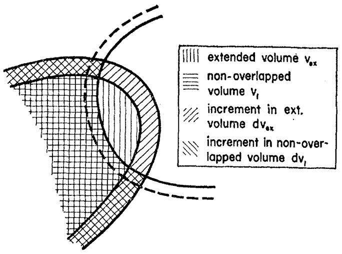

FIG. 8. Figure demonstrating the extended volume V_{ex}, the nonoverlapped volume v_1, and the increments dv_{ex} and dv_1. The intersection of these volumes with the electrodes is shown here.

that part of the extended volume of the domain which is not in common with the extended volume of any other domain (Fig. 8). Let V be the fraction of the total volume occupied by the reversed domains; let dv_1 be the *increment* of the nonoverlapped volume of a domain in a time dt, and dv_{ex} the increment of the extended volume of the domain (Fig. 8). According to Avrami[29] we find the following relation:

$$dv_1/dv_{\mathrm{ex}}=1-\theta.$$

If we denote with $d\sigma_1$, and $d\sigma_{\mathrm{ex}}$ the areas intersected on the surface of the sample by the volumes dv_1 and dv_{ex}, we have for our two-dimensional case:

$$d\sigma_1/d\sigma_{\mathrm{ex}}=1-\theta;$$

and since $Nd\sigma_{\mathrm{ex}}=dA_n$, $Nd\sigma_1=d\theta_n$, we obtain

$$d\theta_n/dA_n=1-\theta. \tag{27}$$

If we assume that in the areas which are not yet occupied by an existing domain the nuclei are formed with a constant probability R per unit time, we have for the number dN formed in a time dt:

$$dN/N_0(1-\theta)=Rdt. \tag{28}$$

Since πr_c^2 is the area occupied by each nucleus at its birth, we have

$$\theta^*=\pi r_c^2N/T,\quad d\theta^*=(\pi r_c^2/T)dN.$$

Consequently, multiplying both sides of Eq. (28) by $\pi r_c^2/T$, we obtain

$$d\theta^*/N_0(1-\theta)=(\pi r_c^2/T)Rdt. \tag{29}$$

Rearranging and summing Eqs. (27) and (29), one finds

$$(d\theta_n+d\theta^*)/(1-\theta)=d\theta/(1-\theta)=(\pi r_c^2N_0R/T)dt+dA_n,$$

from which it follows that

$$\theta=1-\exp\{-[A_n+\pi r_c^2N_0Rt/T]\},$$

and, taking Eq. (26) into account,

$$\theta=1-\exp\{-[A+\pi r_c^2N_0Rt/T-(\pi r_c^2/T)N(t)]\}. \tag{30}$$

Substituting Eqs. (8) into Eq. (30), we have

$$\theta=1-\exp\left\{\frac{-2N_0v_0^2}{R^2T}\left[\frac{R^2}{2}(t+t_0)^2 -R(t+t_0)+1-e^{-Rt}(1-Rt_0+\tfrac{1}{2}R^2t_0^2)\right] +\frac{\pi r_c^2N_0}{T}(1-e^{-Rt})-\frac{\pi r_c^2N_0Rt}{T}\right\}. \tag{31}$$

One can easily show, recalling Eqs. (3) and (4), that both in the cases of true and of apparent sideways motion we can write

$$v_0^2t_0^2=\pi r_c^2. \tag{32}$$

Substituting Eq. (32) into Eq. (31), one finds

$$\theta=1-\exp\{-C[1-R(t+t_0)+\tfrac{1}{2}R^2(t+t_0)^2 -e^{-Rt}(1-Rt_0)-\tfrac{1}{2}R^2t_0^2(1-Rt)]\}, \tag{33}$$

where

$$C=2N_0v_0^2/TR^2.$$

Assuming that a nucleus can be formed anywhere on the electroded surface of the crystal, one has $N_0\pi r_c^2=T$ and hence the coefficient C can, with Eq. (32), be written as

$$C=2v_0^2/\pi r_c^2R^2=2/R^2t_0^2.$$

Let us now introduce $k=1/Rt_0$ and $\tau=Rt$. Equation (33) can now be written:

$$\theta(\tau)=1-\exp\{-2k^2[1-(\tau+k^{-1})+\tfrac{1}{2}(\tau+k^{-1})^2 -e^{-\tau}(1-k^{-1})-\tfrac{1}{2}k^{-2}(1-\tau)]\}. \tag{34}$$

Equations (31) and (34) are equal to Eqs. (9) and (10).

ACKNOWLEDGMENT

We wish to thank Dr. W. J. Merz for many helpful discussions.