Thus at high fields we get

$$\mu/\mu_0 = 1.078 y^{-\frac{1}{4}}, \tag{11}$$

$$\mu'/\mu_0 = 1.143 y^{-\frac{1}{4}}, \tag{12}$$

$$\mu'/\mu = 1.061. \tag{13}$$

CALCULATIONS

The authors have calculated μ/μ_0 and μ'/μ_0 for $y=1, 2, 3, 4, 5, 6, 7, 8, 9, 10$, and 20 by exact integration and for $y=0.1, 0.2, 0.3, 0.5, 0.8$, and 1.0 by numerical integration. The values for $y=1$ obtained by numerical integration and exact integration are found to be in agreement to the third decimal place.

The high-field approximation gives an error of 7% in the values of μ/μ_0 and μ'/μ_0 at $y=20$. The values of μ/μ_0 and μ'/μ_0 obtained by the low-field approximation are 0.939 and 1.020, compared to 0.953 and 1.098 given by the numerical integration for $y=0.1$. Thus the high-field and low-field approximations are justified in the ranges $20<y<\infty$ and $0<y<0.1$ respectively.

The results are graphically illustrated by Figs. 1 and 2.

ACKNOWLEDGMENTS

The authors are grateful to Professor R. E. Burgess for suggesting the problem and helpful discussions. They are also indebted to Dean G. M. Shrum for kind encouragement. Thanks are also due to National Research Council of Canada for award of a fellowship (M.S.S.) and Studentship (P.C.E.) to authors.

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Barkhausen Pulses in Barium Titanate

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A study has been made of the Barkhausen pulses that occur during polarization reversal in single crystals of barium titanate. By both pulse counting and oscillographic techniques, the pulse shapes and in particular their heights and rise times have been studied as a function of the crystal thickness and the applied field strength. The pulse shape represents an initial rapid increase in the volume of the region switched followed by a slower relaxational type of growth, the latter being described by a time constant of 5 to 6 μsec. The pulse heights increase with the crystal thickness and linearly with the applied field while they are practically independent of temperature between room temperature and 94°C. The relaxation time is essentially independent of the crystal thickness, of the applied field, and of the pulse height. The total number of pulses in a given crystal is independent of the field and temperature. In crystals 5×10^{-3} cm thick, the average volume corresponding to a pulse is 10^{-11} cm^3 while the total volume represented by all the pulses is less than one percent of the crystal volume between the electrodes. Individual pulses occur quite independently of each other and of their surroundings.

These observations are not consistent with the usual jerky domain-wall motion models for the generation of Barkhausen pulses. It is concluded that the eventual size and shape of the rapidly switching region represented by a Barkhausen pulse are mainly determined by the crystal thickness and the condition that the depolarizing field within the region must not exceed the applied field. This criterion is successful in accounting for some of the features of the pulses if the region is assumed to be spike-shaped and extending more or less through the crystal thickness, in particular, the average pulse size and its dependence on the field. These deductions suggest that the Barkhausen pulses could represent the nucleation and initial stages of growth of new spike-shaped domains extending along the c axis and that the fixed number of pulses given by a crystal would then indicate a definite number of nucleating sites on the crystal surfaces. Under certain conditions a spike-shaped critical nucleus is consistent with the empirically determined nucleation probability factor, $\exp(-\alpha/E)$, where E is the applied field strength.

To account for the polarization reversal in the remainder of the crystal it is presumed that, after their formation, the spikes expand radially (sideways) in all directions. By using this model the rate of polarization reversal as a function of time can be formulated, assuming that the radial wall velocity is proportional to the field and the nucleations occur randomly. Satisfactory agreement with experiment is obtained at low fields if it is assumed that the expanding domains stop short of overrunning adjacent nucleating sites. Relaxing this restriction for higher fields again leads to good agreement with experiment. Also, the observed dependence of the switching time and the maximum current on the applied field is predicted by using certain approximations.

INTRODUCTION

FROM the investigations made particularly by Merz[1] and Little[2] using electrical and optical techniques has evolved the following description of the sequence by which the reversal of the polarization of barium titanate is accomplished: spike- or wedge-shaped domains are nucleated at the surfaces of the crystals and grow in the forwards direction, that is, along the c axis. Merz concluded that there was negligible sideways expansion of these domains but this is contradicted to some extent by Little's work, though the latter used a quite different orientation of the field with respect to the crystal axes and the direction of the spontaneous polarization.

When the polarization state is slowly reversed many

[1] W. J. Merz, Phys. Rev. **95**, 690 (1954).
[2] E. Little, Phys. Rev. **98**, 978 (1955).

Barkhausen pulses can be resolved.[3,4] The shapes of these pulses are strikingly similar both in a given crystal and among various samples of different origins. In magnetic materials, Barkhausen pulses are usually ascribed to jerky motion of domain walls caused by nonmagnetic inclusions. However, such a process would be expected to result in pulses of varying shapes (reflecting a distribution of inclusion shapes and sizes) which is not the case in barium titanate. Furthermore, preliminary estimates of the size of the region represented by the average Barkhausen pulse in barium titanate were not very different from the domain sizes determined by Merz, suggesting that a careful study of Barkhausen pulses in ferroelectrics might yield information concerning the formation of new domains.

EXPERIMENTAL

To obtain reproducible data it was necessary to cycle the voltage applied to the crystal in a regular manner; studies of the effect of different voltage cycles have been made and these suggest that relaxing space-charge fields present in the barium titanate crystals affect the rate of nucleation of new domains. These experiments together with a description of the circuitry employed will be described elsewhere.[5] In the present studies, steady or slowly rising voltages were applied to the crystal for a definite time interval to produce the Barkhausen pulses followed by a shorter application of a somewhat greater field in the opposite direction to prepare the crystal for the next cycle. At all times while Barkhausen pulses were being studied, the switching speed was kept low enough to prevent superposition of pulses. Two methods of studying and recording the pulses were employed: pulse-height analyses could be made by conventional counting techniques while individual pulses could be studied from photographs of triggered oscilloscope traces. By applying sine waves of suitable amplitude to the amplifier input, it was established that the pulse-height analyzer triggered reliably at frequencies ranging from those corresponding to the fastest rising to those corresponding to the most slowly rising Barkhausen pulses.

The crystals used in these experiments and the

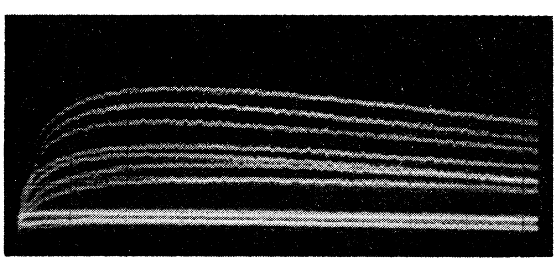

FIG. 1. Oscilloscope photographs of typical Barkhausen pulses. The total length of the trace is 100 μsec.

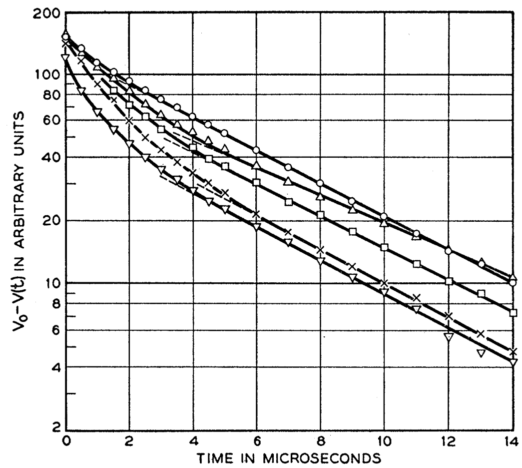

FIG. 2. Plot of $[V_0-V(t)]$ against time where V_0 is the height a Barkhausen pulse would eventually attain in the absence of any distortion due to the input circuit and $V(t)$ is its height at time t.

method of attachment of the electrodes are described elsewhere.[5] The crystals were kept in a dry atmosphere during all the experiments. Examination with a polarizing microscope showed the crystals to be entirely c domained.

RESULTS

I. Study of Pulse Shapes

The growth of a Barkhausen pulse was found to be significant for a period of up to 40 μsec or more. This necessitated the use of input and amplifier circuits with good low-frequency response if appreciable distortion was to be avoided. A photograph of typical output pulses obtained with such a system is shown in Fig. 1. It will be observed that the pulses grow rapidly at first followed by a period where the growth gradually slows down. Analysis of such wave forms showed quite definitely that the output pulse could not result from the effect of the circuit on a linearly rising input pulse (as could result from steady wall motion alone). Nor could it result solely from an exponentially growing input signal of the form: $q=q_0[1-\exp(-t/\tau)]$, with τ some characteristic relaxation time for the pulse and q, its amplitude. In particular, matching the maximum pulse height given by this model to that of the Barkhausen pulse showed that the latter rose initially very much faster than the exponentially growing signal.

The pulse shapes were analyzed in the following manner: an estimate was made of the voltage asymptote, V_0, towards which the output pulse would approach in the absence of any clipping effect due to the circuit. (In practice, V_0 was taken to be slightly higher than the maximum height attained.) Then semilog plots were made of $[V_0-V(t)]$ against t; some typical results are shown in Fig. 2. It is evident that after the initial rapid increase in q, the slower growth can be represented satisfactorily by an exponential term. Thus

[3] Newton, Ahearn, and McKay, Phys. Rev. **75**, 103 (1949).
[4] A. C. Kibblewhite, Proc. Inst. Elec. Engrs. **102B**, 59 (1955).
[5] A. G. Chynoweth (to be published).

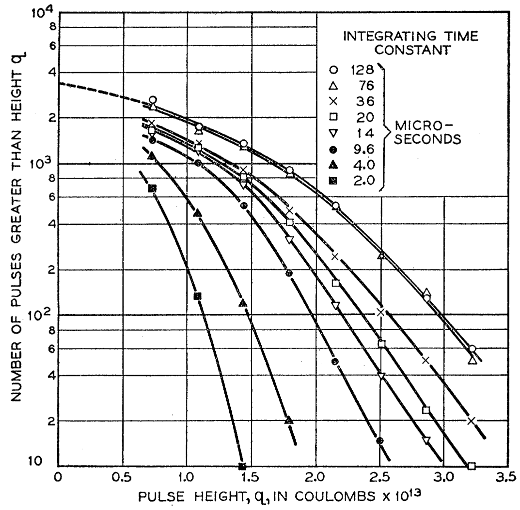

FIG. 3. Integral pulse-height distribution curves for different values of the time constant of the input circuit.

the total pulse shape can be represented approximately by

$$q=(q_1/t_1)t \quad \text{for} \quad 0\leq t\leq t_1,$$

t_1 being the duration of the linear growth, and

$$q=q_1+(q_0-q_1)\{1-\exp[-(t-t_1)/\tau]\} \quad \text{for} \quad t\geq t_1.$$

It should be noted that the distortion to the exponential part of the pulse produced by the circuit now applies mainly to the amplitude (q_0-q_1) and so is relatively much less than when it affects the total amplitude, q_0, if (q_0-q_1) is not very large compared to q_0. From the slopes of the linear portions in the semilog plots, the relaxation time τ was found to be 5 ± 1 μsec. It is also apparent that the amplitude of the rapid first stage is comparable to that of the slower second stage in the growth of the pulse.

The slowly growing part of the pulse was also analyzed by a different technique. The incoming pulse, besides being applied to the Y amplifier of a wideband oscilloscope, was used to trigger a square-pulse generator. The generated pulse, of rise time of about 0.1 μsec and about 50 μsec in duration, was distorted by an integrating RC circuit of variable time constant. The resulting exponentially growing voltage was applied to the X axis of the oscilloscope. The value of RC was adjusted until the oscilloscope traces produced by the pulses, after the initial rapidly rising portion, appeared as near to straight lines as possible. Very good straight lines could be obtained, the corresponding value of RC at typical fields was 6 ± 1 μsec. It is interesting to note that a relaxation time of 5.5 μsec has been deduced from studies of the frequency dependence of the dielectric constant and loss of barium titanate.[6]

[6] Drougard, Funk, and Young, J. Appl. Phys. 25, 1166 (1954).

II. Pulse-Height Analyses

1. *Relation between Height and Rise Time of Pulses*

The pulse rise time is defined arbitrarily as the time taken for the output signal to attain its maximum value. The rise time is thus determined by a combination of the circuit parameters and the shape of the Barkhausen pulse. As described above, the shapes of the pulses could be described quite well by a rapid linear rise followed by a much slower growth, where the time dependence of the slow growth was approximately of the form $[1-\exp(-t/\tau)]$. If such a pulse shape is impressed on an RC circuit of time constant τ_I, then the output pulse has the form:

$$\left(\frac{\tau_I}{\tau_I-\tau}\right)[\epsilon^{-t/\tau_I}-\epsilon^{-t/\tau}].$$

It is easily shown that for $\tau_I\gg\tau$, the output pulse attains a maximum amplitude at time

$$t_R\approx\tau\ln(\tau_I/\tau).$$

Hence, because τ varies more rapidly than the logarithmic term, t_R will increase with τ and therefore, a measurement of the rise time, t_R, does indeed reflect the duration of the slowly growing part of the pulse.

In general, τ_I was kept very large compared with τ but for the measurements described in this section, the integrating time constant, τ_I, of the input circuit was varied by altering the crystal load resistor. Applying square voltage pulses to a 2.5-$\mu\mu$f calibration condenser in the input circuit enabled τ_I to be measured from the resulting waveform displayed on the oscilloscope with calibrated sweeps. Integral pulse height distribution curves were taken for different values of τ_I. A typical set of results is shown in Fig. 3 where the same steady externally applied field was used for all the curves.

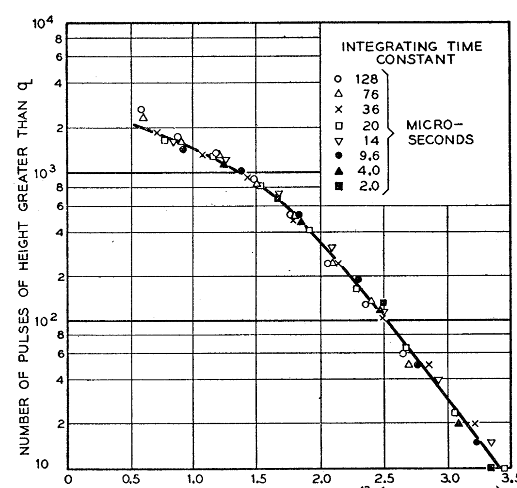

FIG. 4. Normalized integral pulse-height distribution curve obtained by scaling only the abscissas of the curves of Fig. 3.

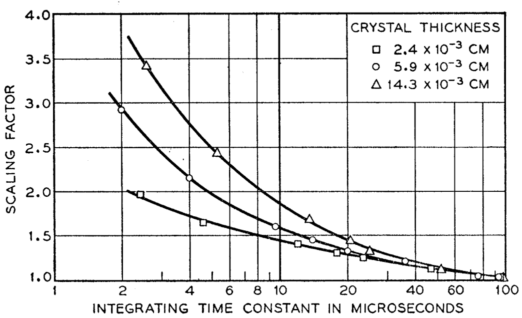

FIG. 5. Plots of scaling factors *versus* input circuit time constant for crystals of different thicknesses. The scaling factor is the amount by which the abscissas of the points on the curve for a particular value of the time constant must be multiplied so that they lie on the curve that applies for infinite time constant, the latter curve being obtained by extrapolation.

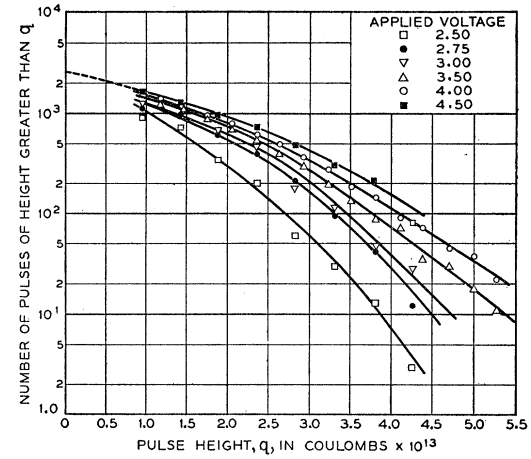

FIG. 6. Integral pulse-height distribution curves for various values of the applied field. (Crystal thickness was 5×10^{-3} cm.)

Qualitatively similar results were obtained for several crystals, most of them showing curves of comparable shape at high values of τ_I. For the larger pulse heights, the count tends towards an exponential drop-off with pulse height while the slope of the curve decreases somewhat at lower pulse heights. As τ_I is reduced, the count at a particular bias drops because of the drastic clipping effect the integrating time constant has on the pulse shapes. Of particular significance, however, was the fact that by choosing suitable scaling factors for the abscissas only, all of the curves could be superimposed to within the limits of experimental error. The result of scaling the curves of Fig. 3 so as to superimpose on the curve for $\tau_I=36$ μsec is shown in Fig. 4.

To interpret this result it is necessary to consider the effect of the input circuit on pulses of different heights, different rates of rise, and different rise times. It will be supposed that, as suggested by inspection of Fig. 1, the pulses have somewhat similar shapes; that is, they can be made to superimpose roughly on one another by a suitable choice of scaling factors for both their magnitude and time axes.

In case (i), suppose that all the pulses have comparable rise times but that there is a distribution of pulse heights. Then, the input circuit will reduce the heights of all the pulses by equal fractional amounts. Thus, the bias curves for different input time constants can be superimposed by a suitable choice of scaling factors for the output pulse heights, in agreement with the experimental situation.

In case (ii), suppose all the pulses have comparable rates of rise but that there is a distribution in rise times, and hence, pulse heights also. Then the input circuit will produce a greater fractional reduction in the height of the pulse the greater its rise time. Thus, as τ_I is reduced, the larger pulses tend to drop out of the bias curves more rapidly than the smaller ones. This leads to a change in the shape of the bias curve, making fitting impossible.

In case (iii), let all the pulses be of the same height but of different rise times. Then, those pulses with longest rise times will suffer greater clipping than those of shorter rise times. Consequently, the input circuit will alter the distribution of pulse heights, so making the fitting of the curves impossible.

Finally, the general case, (iv), where there is a distribution of both pulse heights and rise times, can be regarded as a combination of cases (ii) and (iii) and again, the curve fitting will not be possible.

Thus, it is concluded that most of the actual Barkhausen pulses at a given field have roughly equal rise times and that they differ from each other by a scaling factor for their heights. In particular, the pulse height is independent of the rise time.

By plotting the scaling factors against τ_I it was possible to obtain the bias curve for $\tau_I=\infty$ by extrapolation. In practice, this was seldom very different from the curves obtained at the highest value of τ_I.

2. *Dependence of Rise Time on Crystal Thickness*

In Fig. 5, the scaling factor is plotted against τ_I for crystals of different thicknesses. Here the scaling factor

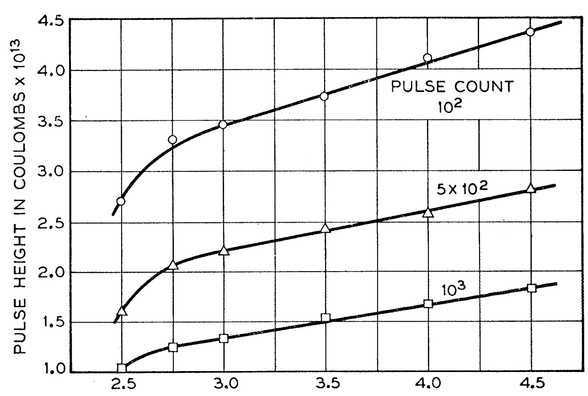

FIG. 7. Dependence of pulse height on field strength as interpolated from the curves of Fig. 6.

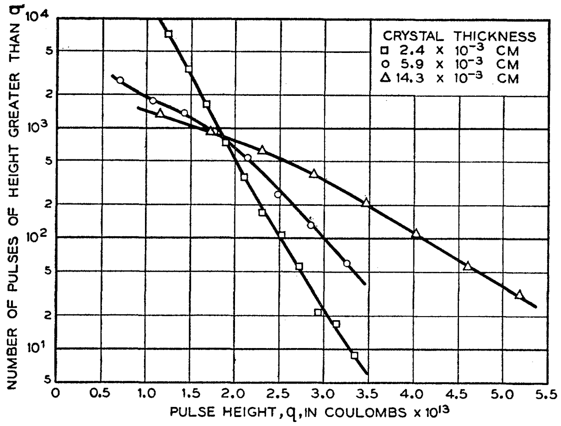

FIG. 8. Integral pulse-height distribution curves for crystals of various thicknesses.

is the amount by which the abscissas of the points on the curve for a particular value of τ_I must be multiplied so that they lie on the curve for $\tau_I=\infty$. Thus, while the scaling factor approximates unity, the input circuit is producing negligible distortion of the pulse shapes. From Fig. 5 it is evident that as τ_I is reduced, pulse-shape distortion sets in first for the thickest crystal and last for the thinnest crystal. This means that the average rise time of the Barkhausen pulses increases with crystal thickness. However, though the semi-logarithmic form of plotting establishes the trend of the thickness dependence of the rise time, it is not easy to determine the magnitude of the dependence. Actually, the five-fold increase in thickness resulted in about a 20% increase in rise time. The data are too meager to allow any more precise conclusion to be drawn.

3. *Dependence of Pulse Height on the Applied Field*

Integral bias curves were taken for different field strengths applied to the crystal; the results are shown in Fig. 6. The range in field strengths is limited by the fact that if the field is too small, pulses occur too infrequently while at somewhat higher fields, the pulses occur too rapidly to be resolved. However, it can be seen from Fig. 6 that all the bias curves appear to be heading for the same intercept on the pulse count axis to within the limits of experimental error. Also, the higher the field, the wider the distribution in pulse heights. Thus, from these data it is concluded that the total number of Barkhausen pulses of all sizes does not vary with the applied field over the given range but that their height increases with the field. Plots of the pulse height *versus* the field as interpolated from Fig. 6 show that the pulse height increases approximately linearly with the applied field (Fig. 7).

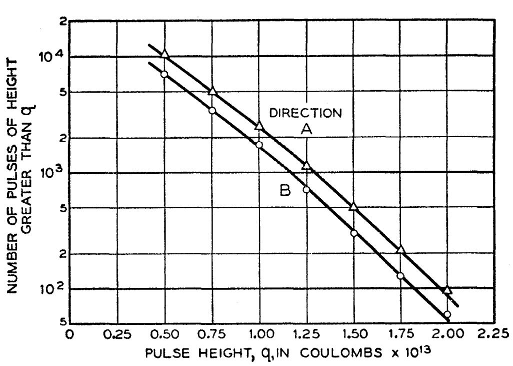

FIG. 9. An example of the pulse-height distribution curves for both directions of the field.

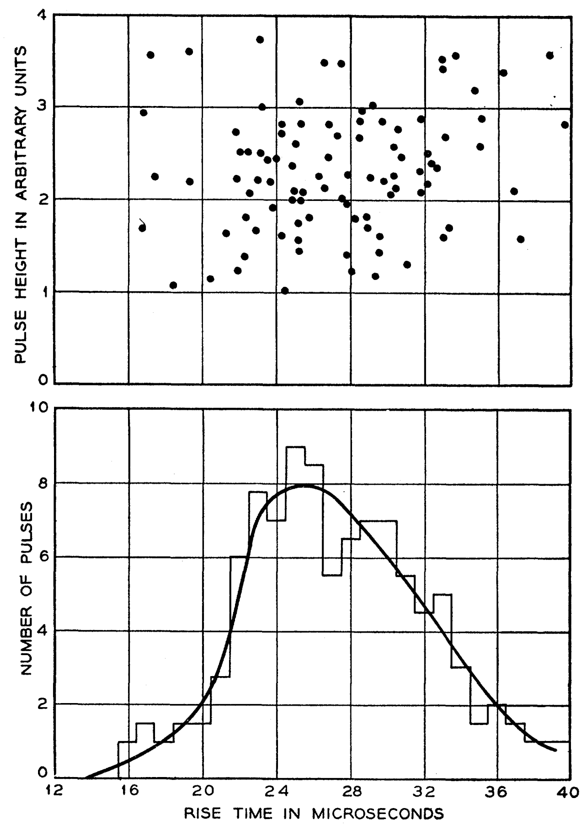

FIG. 10. Top. The points represent the pulse height and rise times for a number of pulses. These points are plotted in the form of a histogram in the bottom figure.

4. *Dependence of Pulse Height on Crystal Thickness*

In Fig. 8, are shown integral pulse count curves for crystals of different thicknesses though of similar electrode areas and with roughly comparable applied fields. As the number of pulses per unit electrode area varied from crystal to crystal it was not possible to make a truly realistic normalization of the ordinates of these curves. With this caution, the results suggest that the pulse height increases with crystal thickness and this was confirmed by qualitative observations on many crystals. Also, it appears that the total number of pulses decreases as the thickness increases though this result was not true of all crystals.

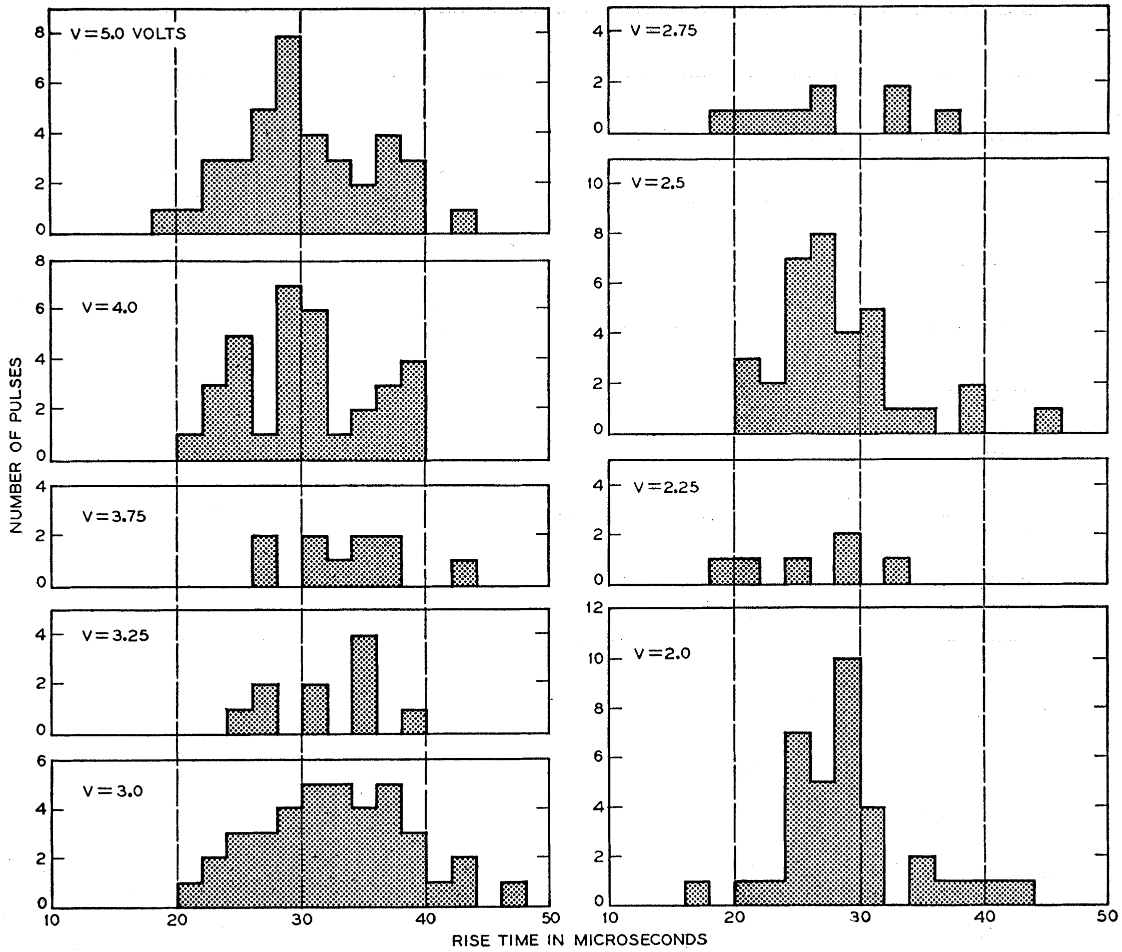

FIG. 11. Rise-time histograms for various values of the applied field.

5. *Effect of Field Polarity*

An example of the bias curves obtained for the two polarities of the field is shown in Fig. 9. In spite of care being taken to ensure that the fields and calibrations were the same, there remained a small but definite disparity between the two curves. It can be concluded that the total number of Barkhausen pulses depends somewhat on the direction of the field. In some crystals there were considerable differences between the two bias curves.

III. Photographic Pulse Recording

1. *Relation between Pulse Height and Rise Time*

Photographs such as Fig. 1 were taken throughout the switching period and from them the rise times and pulse heights were obtained for an appreciable number of pulses, the rise time again being taken as the time required for the output pulse to reach its maximum value, the latter being recorded as the pulse height. The rise time is plotted against the pulse height in the top part of Fig. 10 and it is clear that the points do not follow any particular trend, i.e., it can again be concluded that the rise time of a given pulse bears no relation to its height, which confirms the conclusion drawn from the bias curve measurements described above. Figure 10 shows again that there is an average rise time under the conditions of the measurements. This average rise time can be determined from the histogram shown in the bottom part of Fig. 10. It is also significant that an approximate lower limit can be placed on the rise times.

2. *Dependence of Rise Time on Applied Field*

From histograms showing the rise-time distributions for different applied voltages, the field dependence of the average rise time can be determined. A set of such histograms is shown in Fig. 11, where the "number of pulses" plotted as ordinate refers to the number of pulses photographed and bears no relation to the total number of Barkhausen pulses that occur. The photographs were taken at intervals throughout a switching sequence for each value of the applied field. It is

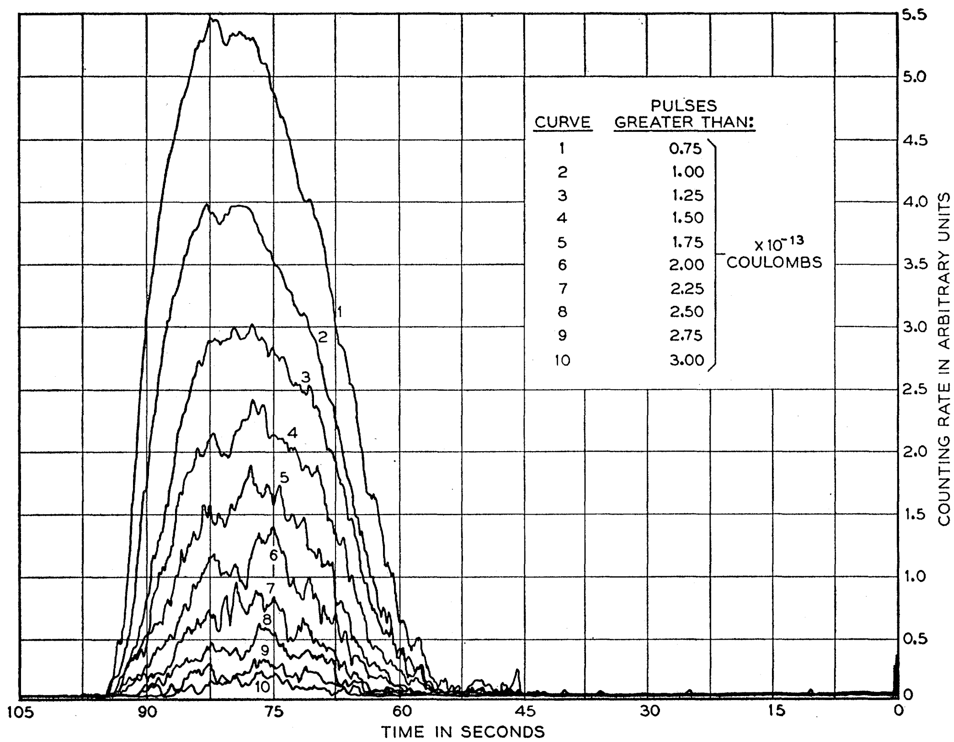

FIG. 12. Barkhausen pulse counting rate *versus* time while the crystal is being switched by a slowly rising field. The different curves apply to various settings of the pulse-height analyzer bias level. The curves are traced directly from a chart recorder.

apparent that there is no appreciable change in the rise time for a field variation by a factor of 2.5.

IV. Counting Rate Studies

1. *Stage of Switching at which the Various Sizes of Pulses Appear*

Using a ratemeter of sufficiently short integrating time constant, the counting rate was recorded on a paper recorder as a function of the time during the switching. The curves so obtained at different settings of the pulse-height analyzer bias level are shown in Fig. 12 as traced directly from the chart record. (For these measurements, a slowly rising voltage was used to switch the crystal, which resulted in a more rapid completion of the switching than that caused by the usual steady voltage.) The lowest curves represent solely the largest pulses while, as the bias level is lowered, the curves move progressively higher reflecting the increasing number of smaller pulses included in the recorded counting rate. From several such experiments it was established that there was no leaning in the set of curves, showing that pulses of all sizes are distributed in the same way throughout the switching sequence.

2. *Counting Rate Versus Switching Current*

By connecting an electrometer across the load resistor in the crystal circuit, the switching current could be studied simultaneously with the counting rate. The electrometer and ratemeter outputs were compared directly by feeding them on to an X-Y recorder. During the switching the recorder pen made an excursion out from its starting point, returning when both signals had again reached zero. A typical trace is shown in Fig. 13; the noisiness of the trace results from the short ratemeter integrating time constant that was necessary to eliminate the lag in the output signal. However, it is apparent that at the field used, the counting rate varied close to linearly with the switching current. In particular, there was no noticeable tendency for the Barkhausen pulses to occur somewhat ahead of the main switching current, this being found to be true over most of the fields used in the present experiments. On the other hand, recent more detailed investigations by Miller[7] have revealed some instances where, with a low field applied to the crystal, a significant number of the pulses do not appear until most of the switching represented by the current has ended.

V. Some Quantitative Results

1. *Maximum and Average Pulse Sizes*

The maximum and average pulse sizes, q_M and $\bar{q}$ respectively, can be estimated from the bias curves taken with the maximum value of τ_I; negligible dis-

[7] R. C. Miller (private communication).

tortion is then introduced by the input circuit. Reasonable values appear to be for q_M, about 4×10^{-13} coulomb, and for $\bar{q}$ about 10^{-13} coulomb for the crystal used to obtain the curves of Fig. 3. The pulse height[3] is given by $q=2Pv/d$, where P is the spontaneous polarization (26×10^{-6} coulomb cm^{-2}), d is the crystal thickness ($\sim5\times10^{-3}$ cm), and v is the volume of the crystal that switches to produce a pulse of charge q. Thus the volumes v_M, $\bar{v}$ corresponding to q_M, $\bar{q}$ respectively, are 4×10^{-11} cm^3 and 10^{-11} cm^3. Using the same argument, Newton, Ahearn, and McKay[3] arrived at volumes about 100 times greater than these. However, their crystal was 30 times thicker than the present one, which supports the above conclusion that the pulse size increases with the crystal thickness.

Kibblewhite[4] has suggested that the above method of estimating the volume associated with a Barkhausen pulse may be fallacious in that it assumes that the usual charge calibration of the input circuit (carried out with the crystal polarization saturated) is the same as that while it is in the midst of switching. To test this assumption the calibration pulse generator was triggered by a

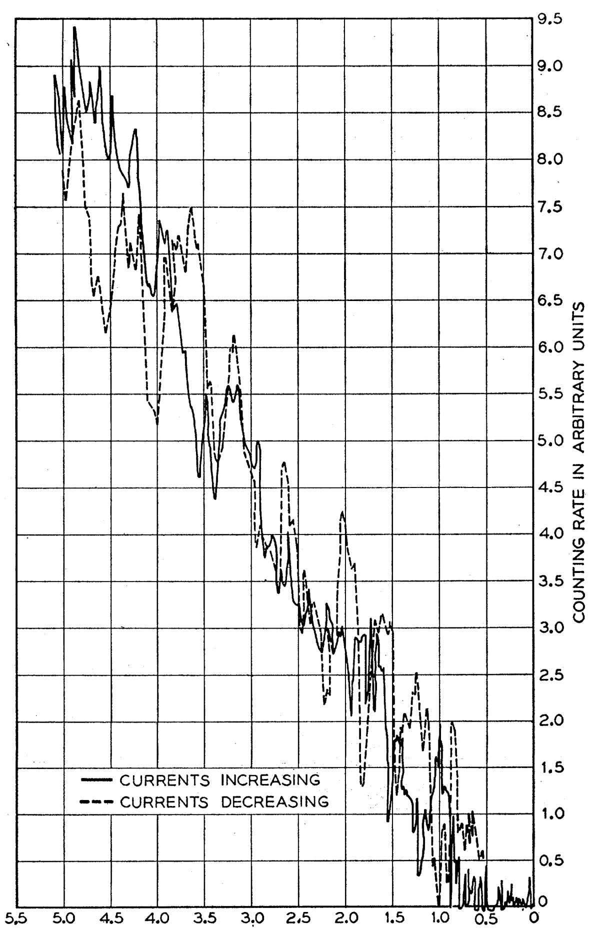

FIG. 13. Barkhausen pulse counting rate *versus* switching current as traced by the X-Y chart recorder.

Barkhausen pulse so that the oscilloscope showed the calibration pulse (of about 10 μsec duration) superimposed on the rising part of the Barkhausen pulse. No difference could be detected between the amplitude of the calibration pulse in the two methods. To obtain an estimate for v, Kibblewhite assumed that the total charge in the Barkhausen pulses at high enough fields was equal to the total charge switched by the crystal, $2PA$, and thus, knowing the total number of pulses, he arrived at a value for v. Kibblewhite estimated the total number of pulses at high fields by interpolating data obtained at low fields and at a pulse-height analyzer bias level other than zero. The evidence of Fig. 6 indicates that it is misleading to extrapolate the low-field pulse count at a bias level other than zero to other field strengths, particularly for fields greater by an order of magnitude.

The area under the differentiated bias curve of Fig. 3 for $\tau_I=128$ μsec represents the total charge, Q, contained in the pulses. Graphical integration showed that Q was about 0.1% of the total charge $2PA$. The value of this fraction varied somewhat from crystal to crystal though it was usually much less than one percent. These results are in good agreement with that of 0.4% obtained by Newton, Ahearn, and McKay at low fields while their estimate of about 6% at high fields may be at fault because of the danger of extrapolating low-field data to high fields without knowing the pulse-height dependence on the field.

VI. Effect of Temperature on Barkhausen Pulses

The effect of temperature on the Barkhausen pulses was studied by obtaining bias curves at various temperatures, the field being kept the same. Since increasing the temperature produced faster switching the field was made just sufficient to switch the entire crystal at room temperature within the time that the field was applied. In this way it was possible to obtain runs quite close to the Curie point without the switching being so rapid as to produce pulse pile-up. The input time constant was high so as to avoid distortion of the pulses. As the temperature altered, so did the crystal capacity, thereby necessitating separate charge calibrations of the input circuit for each of the temperatures at which bias runs were made. The results are shown in Fig. 14 and it is clear that to within experimental error, the total number of pulses, N_0, is constant, and also, the pulse height is independent of the temperature.

VII. Abnormal Pulse Shapes

The complex pulse shapes noted by previous authors[3] were rarely seen in the present studies. An interpretation of these pulses is that they represent the triggering of a nucleation by another through the agency of a-domain coupling; a domains were present in Kibblewhite's crystals and probably in those of Newton, Ahearn, and

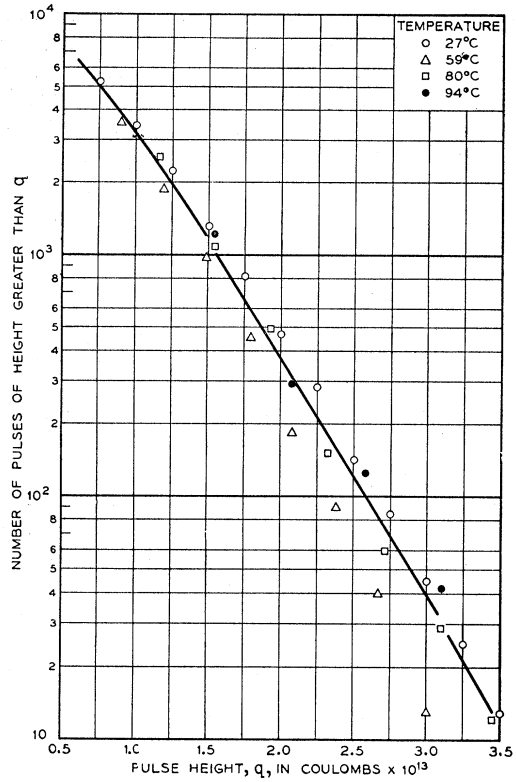

FIG. 14. Integral pulse-height distribution curves at various temperatures.

McKay also, whereas the crystals described in this paper were free from a domains.

Abnormal pulse shapes were seen occasionally, however. Figure 15 shows some typical examples of pulses which rise slowly at first but faster than linearly with time.

VIII. Summary of Experimental Results

Let t_R represent the average rise time for the group of pulses produced throughout the switching sequence and let N_0 be the total number of pulses as extrapolated from the bias curves. Let q represent the pulse height and let d represent the crystal thickness and V the applied voltage. Then, the above experiments have shown that:

(i) At a given field, there is no relation between t_R and q for a given pulse.

(ii) t_R increases slightly with d.

(iii) There is an appreciable minimum value of t_R under given switching conditions, there being no pulses with rise times shorter than this.

(iv) The average and maximum values of q increase with d.

(v) q increases linearly with V.

(vi) N_0 remains constant while V is varied.

(vii) N_0 appears to increase as d decreases.

(viii) N_0 differs with the polarity of V.

(ix) There is no evidence for pulses of any given size showing any preference for a particular period during the switching cycle.

(x) The pulses occur at a rate proportional to the total switching current (except at low V when they sometimes occur towards the end of the switching current).

(xi) The maximum and average-sized pulses correspond to the switching of crystal volumes of 4×10^{-11} cm^3 and 10^{-11} cm^3, respectively, in crystals of thickness 5×10^{-3} cm.

(xii) The total charge obtained by summing the charges in all the individual pulses is less than one percent of the total charge switched in the crystal.

(xiii) Individual pulses represent, apparently, a two-stage process for the polarization reversal in a small region, an initial rapid change in the polarization followed by a more slowly growing phase. The slow growth can be described by a relaxation time of 5.5 μsec.

(xiv) The number and size of the Barkhausen pulses do not vary with temperature over the range 27°C to 94°C.

It is relevant to include some of the facts about the Barkhausen pulses that will be given in reference 5.

(xv) The rate of appearance of the pulses is influenced by the field in the surface layers of the crystal near the electrodes.

(xvi) The rate of appearance of the pulses is governed by the field, E through a probability factor which, at least, approximates $\exp(-\alpha/E)$, where α is a constant.

(xvii) Individual pulses occur independently of each other and of their surroundings.

INTERPRETATION OF BARKHAUSEN PULSES

I. Jerky Wall Motion Hypothesis

In magnetism, Barkhausen pulses are commonly interpreted as being manifestations of jerky motion of a domain wall.[8] It has been postulated that in magnetic materials there exist nonmagnetic inclusions which hinder the motion of a domain wall.[9] The Barkhausen pulse can arise when the wall, or part of it, snaps past

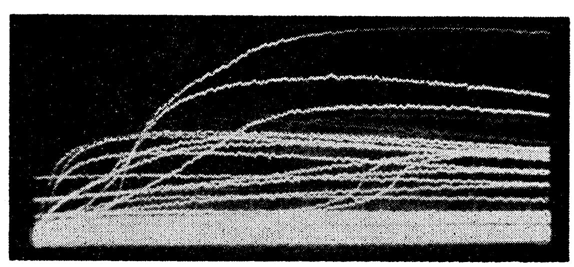

FIG. 15. Oscilloscope photograph showing some of the abnormal pulse shapes that were occasionally observed.

[8] H. J. Williams and W. Shockley, Phys. Rev. **75**, 178 (1949).
[9] R. S. Tebble, Proc. Phys. Soc. (London) **B68**, 1017 (1955).

the inclusion. Alternatively the pulse may be produced by the wall, or part of it, becoming free of the inclusion and sweeping on until it is again stopped. That the pulses in barium titanate whose characteristics are outlined above cannot be accounted for by such mechanisms is the conclusion reached from the arguments detailed below.

In this discussion, two basic assumptions are made, namely:

(i) The inclusion density is roughly the same for all the crystals used (which came from the same melt and were processed in identical ways).

(ii) The velocity of a domain wall varies roughly as the applied field in both the forward and sideways directions—a linear dependence of the wall velocity on the field has been demonstrated in Rochelle salt[10] and also, it is the observed behavior in magnetic materials.[11]

The magnitude of the Barkhausen pulse is determined by the volume of material swept out by the moving wall, that is, q varies as Ax, where A is the wall area and x is the distance it travels.

There are four basic models for jerky wall motion, each of which must be considered for sideways as well as forward motion. Schematic representations of the various models are shown in Fig. 16. Let s represent the linear dimension of an inclusion and r the distance between inclusions.

Model A.—When an inclusion is encountered, the whole wall moves rapidly into a position of equilibrium at the inclusion. Alternatively, if the field is sufficient, the wall can move discontinuously through the inclusion and then proceed at its former smooth and slower pace. The pulse height, q, will be determined by As. There are two subdivisions to this model: (a) the distances s swept out by the wall are roughly the same for all the walls, then the distribution in q arises from a distribution in A; (b) the wall areas are approximately the same, the range in pulse heights representing the range in s.

Model B.—The wall can be regarded as getting trapped at an inclusion, eventually breaking away and moving rapidly until it is trapped by the next inclusion. In this case, $q \propto Ar$. This model can also be subdivided into (a) approximately constant r and (b) roughly the same A for all pulses.

Model C.—Instead of the whole wall behaving rigidly, the motion of only a part of the wall may be affected by the inclusion while the major part of the wall continues to move on. The hindered part produces a pulse by moving discontinuously into or across the inclusion as in model A; the size of the pulse reflects the size of the inclusion.

Model D.—Similar to model C, except that the hindered part produces a pulse when it breaks away from the inclusion and catches up with the rest of the wall. The size of the pulse is determined by the distance that the hindered part has to catch up which in turn, is determined by the time the segment of the wall remained trapped at the inclusion.

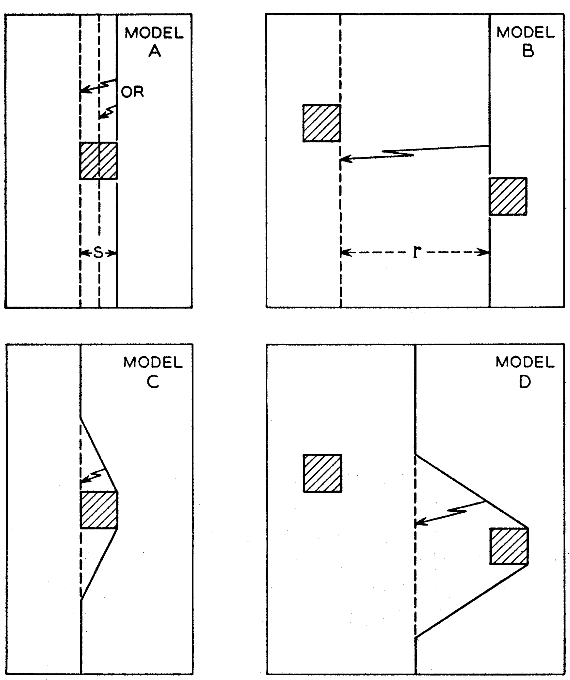

FIG. 16. Diagram illustrating the models for jerky wall motion discussed in the text.

Consideration of the above models leads to various predictions as to the field and thickness dependence of the measured heights of the pulses. These predictions are summarized in Table I. In cases where the distance jerked by the wall in the thickness direction is proportional to the size s or the distance r, the electrostatic situation decrees that the magnitude of the signal depends on the value of (s/d) or (r/d) (line 1). For sideways motion of walls extending through the whole thickness of the crystal, q is independent of d (line 2).

Ignoring the electrostatic problem and considering only the effect of the increasing total number of inclusions as d increases (keeping electrode area constant), then, an extensive wall in forward motion will encounter more inclusions while crossing through the total thickness of the crystal. In sideways motion, though the distance it travels remains the same, increasing d will cause an extensive wall to encounter inclusions more frequently. Thus, N_0 increases with d and this will certainly be true also for all models in which part of the wall is hindered by the inclusion (line 4). In sideways motion, model B requires that q decreases as d increases (line 3).

Keeping d constant and increasing V will have no effect on the sizes of the pulses (line 9) except in the complicated case of model D where the volume swept out by the wall depends on the time it is trapped at the inclusion which, in turn, will be some function of the field.

[10] T. Mitsui and J. Furuichi, Phys. Rev. **95**, 558 (1954).
[11] J. K. Galt, Bell System Tech. J. **33**, 1023 (1954).

TABLE I. Comparison of the observed field and thickness dependence of the pulse heights and rise times with the behavior predicted by the various models for jerky wall motion.

	Model Sideways	Model Forward	Prediction	Experimental
	a. For comparable field strengths, different crystal thicknesses			
1	*C*, *D*	*Aa*, *Ab* *Ba*, *Bb* *C*, *D*	q is inversely proportional to d (by considering the electrostatic situation only and ignoring effect of greater number of inclusions)	q increases with d
2	*Aa*, *Ab* *Ba*, *Bb*		q is independent of d (wall extending through crystal thickness and considering electrostatic problem only)	q increases with d
3	*Ba*, *Bb*		q decreases as d increases (because of greater total number of inclusions and ignoring electrostatic problem)	q increases with d
4	All models		N_0 increases with d	N_0 decreases with d
5	*Ab*, *Bb* *C*, *D*	*Ab*, *Bb* *C*, *D*	q increases with t_R	q independent of t_R
6	*Aa*, *Ba*	*Aa*, *Ba*	q independent of t_R	q independent of t_R
7	All models		t_R independent of d (under same conditions as line 1)	t_R increases with d
8	*Ba*, *Bb*		t_R decreases as d increases (under same conditions as line 2)	t_R increases with d
	b. Crystal of given thickness, various applied fields			
9	*Aa*, *Ab* *Ba*, *Bb* *C*	*Aa*, *Ab* *Ba*, *Bb* *C*	q is independent of V	q increases linearly with V
10	*Aa*, *Ab* *Ba*, *Bb* *C*	*Aa*, *Ab* *Ba*, *Bb* *C*	t_R decreases as V increases	t_R independent of V

It is also possible to make some predictions concerning the field and thickness dependence of the rise time, assuming it to be related to domain wall motion. It will be apparent later, however, that this assumption is, most likely, false. Nevertheless, for the sake of completion, the predictions are included in Table I.

For all the models where the spectrum of pulse heights reflects a spectrum in the distances travelled by the walls it is to be expected that the greater the distance traveled, the longer it takes, that is q increases with t_R (line 5). For those particular models where the spectrum of pulse heights is brought about by a range of wall areas, all walls moving about the same distance, t_R will be the same for all pulses (line 6).

If the field is kept constant, then, for all models, t_R will remain the same while d is varied (considering the electrostatic problem only and ignoring the effect of more inclusions, line 7). If the electrostatic problem is ignored, the increasing number of inclusions as d increases will cause an extensive wall to move shorter distances between inclusions when in sidewise motion. Then, t_R will decrease as d increases (line 8).

Finally, except again for the complex situation of model D, since the distance moved by the wall is independent of V while its velocity increases with V, t_R will decrease as V increases.

On comparing the predictions listed in Table I with the experimentally observed behavior, it is apparent that all of the above models of jerky wall motion fail completely to account for the observed behavior. The same conclusion is reached even if all the predictions about t_R are excluded from the table. Some further considerations follow.

An alternative proposed explanation of Barkhausen pulses is that the more or less steady motion of a domain wall past an inclusion results in sudden rearrangements, in the subsidiary domain pattern surrounding the inclusion.[12] However, this alone would give rise to both positive and negative pulses whereas, in the crystals used for these experiments, only pulses of sign appropriate to the direction of switching were observed. This rearrangement mechanism is of no consequence if it occurs simultaneously with the much larger discontinuities discussed above.

Because there will be a range of inclusion sizes, there will be a range of pulse sizes for the cases where the wall moves discontinuously through the inclusion, i.e., models A and C. It is to be expected that the relaxation time for moving a wall through an inclusion will increase with s, or alternatively, higher fields will be required. Thus, if a slowly rising field is applied to the crystal, the smaller pulses should occur earlier, and the largest ones at the end of the switching. This is quite contrary to the conclusions drawn from Fig. 12.

For model B, the distribution of pulse sizes should show a maximum corresponding to the average distance between inclusions. Differential bias curves show no evidence of any such maximum; rather, the number of pulses of given height drops continuously with height.

If jerky wall motion is responsible for the pulses, the maximum counting rate would be expected to occur near the maximum of the total switching current when, presumably, the greatest number of walls are in motion. Though such a correspondence is represented by Fig. 13, preliminary studies at lower fields have indicated that sometimes very few pulses occur until most of the total charge has been switched,[7] i.e., there may be no correlation between the pulses and the switching current. This phenomenon will be discussed in a future paper. However, it is difficult to conceive of a wall motion that is jerky (owing to energy minima) at high fields becoming less jerky at lower fields.

Conceivably, Barkhausen pulses could be generated if two domain walls in sideways motion approached each other closely, the pulse corresponding to a hastened polarization reversal of the narrowing space between the walls. The expected consequence of this, however, would be for the counting rate to always lag behind the switching current and become a maximum towards the end of the switching.

The arguments outlined above as well as further consideration of the remaining experimental facts listed lead to the conclusion that the Barkhausen pulses are not manifestations of jerky wall motion. It should be

[12] L. F. Bates and D. H. Martin, Proc. Phys. Soc. (London) **B69**, 145 (1956).

noted also that the very great differences in the domain wall thickness of ferroelectrics and ferromagnetics might argue against jerky wall motion models for ferroelectrics.

II. Polarization Reversal in Spike-Shaped Region

The fact that the Barkhausen regions are of finite size and that the variation of the average size with field and crystal thickness follows a well-established pattern suggests that the extent of the Barkhausen region is determined by energy considerations. In this section, the possibility that the Barkhausen region is in the form of a long thin spike extending along the c axis will be explored. Such a picture is made attractive on several counts: (1) The Barkhausen volumes are not very different from those of the smallest spike-shaped nuclei (or domains) observed optically by Merz[1] and Little,[2] and by etching techniques by Hooton and Merz[13] and Cameron.[14] (2) These domains nucleate near the crystal surfaces which is consistent with the conclusion that mobile space charges modulate the field near the electrodes and so regulate the occurrence of the Barkhausen pulses[5] through the nucleation probability factor, $\exp(-\alpha/E)$. (3) The Barkhausen volume is in very close agreement with that calculated by Landauer[15] for a spike-shaped quasi-stable nucleus. (4) If it is supposed that the spike grows mainly in the forwards direction (along the c axis) and that the initial rapid rise of the largest pulses represents motion essentially through most of the crystal thickness, then the forwards wall mobility determined from the Barkhausen pulses is about 3 cm^2 volt^{-1} sec^{-1} (at a field of 600 volts cm^{-1}) which compares very well with that deduced by Merz from switching studies, 2.5 cm^2 volt^{-1} sec^{-1}, and is consistent also with Little's forward velocity measurements.

The change in free energy resulting from the rapid polarization reversal in a given region is usually taken to be the sum of three terms: the electrostatic energy due to the applied field, the electrostatic energy due to the depolarizing field, and the energy due to the domain wall energy. It is of interest to examine whether the main features of the behavior of the Barkhausen pulses can be accounted for by the condition that the depolarizing field within the Barkhausen region must not exceed the applied field. From Landauer's work it follows that this condition can be written (in equilibrium) as

$$\epsilon_a E = 2LP, \tag{1}$$

where E is the applied field and L is the depolarizing factor. The factor 2 appears because of the head-to-head or tail-to-tail configurations of the polarization vectors at the domain boundary and the permittivity along the a axis, ϵ_a, enters as a consequence of the dielectric anisotropy of barium titanate. If the nucleus is approximated by a prolate spheroid of length l and radius r, with $l \gg r$, L is given by,[16]

$$L = (4\pi/m^2)(-1+\ln 2m). \tag{2}$$

In this expression $m = l^*/r$, where $l^* = (\epsilon_a/\epsilon_c)^{\frac{1}{2}} l$, and ϵ_c is the permittivity along the c axis. Using the values quoted by Landauer, $(l^*/l) \simeq 4 \pm 1$.

From Eq. (2) m can be calculated for any value of E. Putting $E = 600$ volt cm^{-1} yields $(l/r) \sim 10$. This estimate is less than the length-to-radius ratio observed by Merz by up to an order of magnitude. A similar discrepancy results if the known Barkhausen volume is taken to represent a spike extending right through the crystal. However, the calculated value of m can be regarded only as very approximate as it pertains to ideal geometry and field configurations (that is, with no image fields due to the presence of an electrode other than the one which nucleated the domain) and there is considerable uncertainty as to the value to be used for ϵ_a. Also, the effect of domain wall energy has been ignored though it can easily be seen that if this were more important than the depolarizing energy, very different field and thickness dependencies would result for the Barkhausen pulses. In particular, an increase in the field would cause a decrease in the Barkhausen region. For a spike-shaped region of volume 10^{-11} cm^3 and $l/r = 10$, it follows that $r \sim 10^{-4}$ and $l \sim 10^{-3}$ cm. This makes the calculated spike extend only a fifth of the way through the crystal whereas in view of the thickness dependence of the average pulse size it seems necessary to suppose that the average spike extends more or less right through the crystal. The calculated basal radius is not inconsistent with the radii of small domains observed optically or by etch patterns. Scatter in the values of l and r could then account for the observed distribution of pulse sizes though it is purely an empirical fact at this stage that frequently the distribution of pulse heights is approximately exponential.

The pulse height q is given by

$$q = 2Pv/d, \tag{3}$$

where v is the volume switched and is proportional to $r^2 l$. From (2) and (3) it follows that

$$q \propto (\epsilon_a^2/\epsilon_c) E l^3/d. \tag{4}$$

Thus, q increases linearly with E, as observed, and it is independent of P. Increasing d may cause the average value of l to increase though probably more slowly. Thus the empirical fact that q increases roughly in proportion to d over the relatively narrow range studied is not inconsistent with (4); for thin crystals it would be expected that l would be limited by d, making $q \sim d^2$, whereas for thick crystals, l may approach a constant making q vary as d^{-1}. The insensitivity of pulse size to

[13] J. A. Hooton and W. J. Merz, Phys. Rev. **98**, 409 (1955).
[14] D. P. Cameron, I.B.M. J. Research and Development **1**, 2 (1957).
[15] R. Landauer, J. Appl. Phys. **28**, 227 (1957).
[16] J. A. Osborn, Phys. Rev. **67**, 351 (1945).

temperature is not explained because of appreciable changes in the permittivities over the temperature range covered. It must be concluded, therefore, that the simple condition that the depolarizing field balances the applied field is only partially successful in accounting for the observed behavior of the Barkhausen pulses.

The fact that there is a marked uniformity in the pulse shapes might also argue in favor of some generally applicable energy criterion but the cause of the actual pulse shape can only be guessed at at present. It seems plausible to associate the initial rapid rise with forward growth of a spike while the slower relaxational growth may represent an adjustment of the spike shape to that of a long, thin, domain extending more or less right through the crystal, the rate of adjustment being controlled by image fields both ahead of and within the switched region. The 5-μsec relaxation time, which seems to be characteristic of barium titanate and independent of its impurity content, would not be particularly evident in high-field switching current transients as the Barkhausen pulses account for less than one percent of the total switched charge. Alternatively, the relaxation time might become negligible at high fields. On the other hand, there is some evidence[17] that the relaxation might be associated with the dynamic behavior of the space-charge layers near the surfaces of the crystal.

It is also pertinent to consider whether the slow growth could result from some sideways growth of the spike due to the applied field. It is not unreasonable to picture sideways motion of a wall as a switching of the dipoles adjacent to the wall so that the switched region propagates basically in the forward direction, layer by layer.[2] Thus, the net field promoting this switching is determined, at least partly, by the applied field, which, in turn, will influence the velocity of the domain wall. It is easy to show that as a consequence, the growth rate will depend in some way on the value of the applied field; this applies equally to wall motion in either the forward or the sideways directions. A further objection to ascribing the slow growth to sideways expansion promoted by the applied field arises from the fact that pulses of various heights have equal rise times; if the different heights reflect to some extent different amounts of sideways travel, the larger pulse heights would be expected to possess larger rise times.

Thus it appears that slow sideways wall motion of the sort envisaged in the previous paragraph does not occur. (Note, though, that it seems necessary to expect some sideways motion to occur in a time not greater than the time taken for the forward growth. This conclusion follows if it is assumed that the effect of the depolarizing field is important during much of the growth of the nucleus.)

If, as seems plausible, a Barkhausen region represents the start of a new domain, it is necessary to reconsider briefly the nucleation problem. From switching studies, Merz[1] deduced that the probability of nucleation was determined by the factor $\exp(-\alpha/E)$ and he could account for the E^{-1} dependence of the exponential factor by allowing the critical nucleus to be wedge-shaped. This restriction is not necessary, however, if the depolarizing field during nucleation is negligible. On this interpretation, the Barkhausen pulse represents the growth of a nucleus *after* it reaches critical size and, consequently, the critical nucleus must be much smaller than the Barkhausen volume. If the critical nucleus is assumed to be in the shape of a prolate spheroid, then its length/radius ratio could be much larger than the value that makes the depolarizing field important. Neglecting the depolarization energy term in the expression for the free energy, it becomes

$$\Delta F = -2EP(\tfrac{2}{3})\pi r^2 l + \sigma(\pi^2/2) r l,$$

where σ is the wall energy. Now if the fluctuations in ΔF are brought about mainly by fluctuations in r, then the condition that $(\partial\Delta F/\partial r)=0$ results in the critical value of r being

$$r^* = 3\pi\sigma/16EP.$$

Thus, the critical free energy is

$$\Delta F^* = (3\pi^3\sigma^2 l/64)/EP,$$

and the nucleation probability is $\exp(-\Delta F^*/kT)$ which, in Merz's notation, is equal to $\exp(-\alpha/E)$. Hence,

$$\alpha = 3\pi^3\sigma^2 l/64PkT.$$

Merz has determined α to be 5×10^3 volts cm^{-1}. Thus, at room temperature,

$$\sigma^2 l = 3.4\times10^{-8}\ \mathrm{erg}^2\ \mathrm{cm}^{-3}.$$

Now for the depolarization field to be unimportant we must have $l/r \gg 10$. Also, at $E=600$ volts cm^{-1},

$$r^* = 3.9\times10^{-6}\sigma.$$

To meet these requirements, σ must be appreciably smaller than about 10^{-1} erg cm^{-2}.

From these estimates, the critical-size nucleus that emerges is a long thin spike a few lattice constants in diameter and extending perhaps for one micron or more into the crystal along the c axis. Thus, the condition that its length be considerably less than the crystal thickness is met. The value that σ is required to have is considerably less than that previously estimated by Anderson[1] (10 erg cm^{-2}), and more recently by Kinase and Takahasi[18] (1.4 erg cm^{-2}), but for such a small critical nucleus, the validity of an estimate based on the macroscopic permittivity and elasticity constants is questionable. Also, it is conceivable that crystal imperfections or dislocations which may provide the nucleating sites could drastically modify the crystal properties in the immediate neighborhood. The lower

[17] Private communication from M. Drougard, International Business Machines, Poughkeepsie, New York.

[18] W. Kinase and H. Takahasi, J. Phys. Soc. Japan 12, 464 (1957).

value of σ arrived at above facilitates sideways motion of domain walls much more so than did Anderson's estimate.

The fact that the total number of Barkhausen pulses produced by a crystal when switched is independent of the applied field and the temperature strongly suggests that nucleations occur at a very definite set of sites at or near the crystal surfaces. It is reasonable to suppose that these nucleating sites lie at crystal imperfections of some sort, possibly dislocations.

The total number of Barkhausen pulses was found to vary over an order of magnitude or more from crystal to crystal of equal electrode areas. In view of the above discussion this could reflect differences in the concentration of surface imperfections amongst crystals. Further study of this point is in progress. Though the number of pulses from a crystal remained constant as long as it was regularly cycled, slow changes in N_0 did occur when the crystal was left for some time. Also, N_0 appeared to depend somewhat on the electrical history of the crystal prior to being regularly cycled. It is unlikely that these changes in N_0 reflect changes in the number of nucleating sites resulting from some sort of annealing or plastic flow occurring at room temperature. Rather it is felt that electrochemical reactions may take place at the surfaces of the crystals, the resulting space charges serving to modify in some way the nucleating properties of the imperfection sites.

Finally, it is of interest to discuss the way in which the whole crystal reverses its polarization in the light of the Barkhausen pulse studies. It has been noted above that the total charge represented by all the Barkhausen pulses amounts to less than one percent of the charge $2PA$. Also, the total switching current is, at all stages of the switching, more or less proportional to the rate of occurrence of Barkhausen pulses. Thus, it seems necessary to conclude that a domain cannot switch until a nucleation has occurred but that the volume of the nucleus corresponding to the Barkhausen pulse is 10^2 to 10^3 times smaller than that of the domain it controls. Thus it appears to be most likely that the Barkhausen volume expands, on the average, by a factor of 10^2 to 10^3 and the most plausible picture is that it does so by sideways motion of the 180° walls. Little's experiments together with studies of etch patterns tend to support this conclusion. The velocity of the sideways motion is limited by the fact that the expansion must take 10^{-1} sec or longer for the height and shape of the Barkhausen pulse to be relatively unaffected. On the other hand, the switching current does not lag the Barkhausen pulses by more than 1 sec. Thus, the expansion of the nucleus by a factor of 10^2 to 10^3 occurs in a time between 10^{-1} and 1 sec. Since the Barkhausen nucleus is regarded as a spike, it will most likely expand radially. The radial velocity of the wall motion is therefore, $10^{-3\pm1}$ cm sec^{-1}. Thus the ratio of forwards to sideways velocity at fields of the order of 10^3 volt cm^{-1} is of the order of $10^{6\pm1}$, a ratio that is not inconsistent with Little's description of the growth of a nucleus.

From the experimental facts it appears that at low fields the expanding nuclei do not overrun adjacent regions that have not been nucleated. This conclusion follows from the independence of N_0 on the field and, particularly, from the exponential tails to the Barkhausen pulse counting-rate curves. It is this last fact which shows that Barkhausen pulses occur independently of each other and of their surroundings. The reason for not ingesting adjacent nucleating sites is not clear. Speculating, as nucleating sites probably reside at crystal imperfections they will be surrounded by stress fields such that a domain wall moving in the stress field of its nucleating site may find a potential barrier preventing it from entering the stress field of an adjacent nucleating site. At high applied fields, the wall may be able to get past the barrier.

Summarizing, the Barkhausen pulse studies have suggested that at low fields, a crystal switches by the random nucleation of a certain number of spike-shaped nuclei which extend through the crystal thickness and thereafter grow by slow sideways expansion until the whole of the crystal is switched though the expanding domains do not overrun adjacent nucleating sites. This model enables the switching current transient to be formulated if some assumption is made as to how the nucleus expands. This is done in the Appendix where it is shown that good agreement with experimentally obtained current transients results for both the low- and the high-field regions.

CONCLUSIONS

From the detailed studies of the electrical characteristics of Barkhausen pulses in barium titanate, it is concluded that they do not represent domain wall jerks of the particular kinds usually felt to be responsible for Barkhausen pulses in magnetic materials. An attempt to account for their properties on the basis of the condition that the depolarizing field in the Barkhausen region must not exceed the applied field is partially successful if the region is assumed to be spike-shaped and lying along the c axis. An alternative condition assuming domain wall energy to be the dominant factor is much less satisfactory. The depolarizing field criterion yields Barkhausen regions not too different from the smallest spike-shaped domains that have been observed optically, suggesting that the Barkhausen pulses are manifestations of the formation of new domains. If this is correct it seems necessary to conclude that after its formation, a spike-shaped domain must expand sideways in all directions with a wall velocity slow compared with the forward growth of the spike. This radial, or sideways, wall motion eventually ceases; it appears that there is a potential barrier preventing an expanding domain from overrunning an adjacent region controlled by another nucleating site. This barrier may arise from crystal strains associated with imperfections that provide the nucleating sites. The volume of a domain when it ceases to grow is of the order of 10^2 to 10^3 times greater than that of the initial spike. When all the nucleations and expansions have occurred, the crystal is fully switched.

Merz's picture of switching is modified to some extent since the Barkhausen experiments indicate that a relatively large amount of sideways wall motion occurs. However, the resulting domains in crystals of the thicknesses normally used will still appear as long columns, circular in cross section. Thus, it is true to say that at low fields, the crystals switch by the appearance of many thin domains rather than by walls sweeping sideways through the crystal in the same way as they do in magnetic materials. The picture of a nucleus first growing rapidly in the forwards direction and then expanding very slowly in the sideways direction is in complete agreement with Little's conclusions.

Formulations of the switching current transient based on the above model of switching lead to pulse shapes which reproduce the main features of the observed transients at low fields. Also, the correct form for the field dependence of the switching time and the maximum current are predicted. At high fields the model becomes modified in that it appears that the sideways moving domain walls can sweep on through the crystal overrunning many nucleating sites before they have had a chance to nucleate. Formulating the switching transient resulting from this model again leads to agreement between the predicted and observed pulse shapes as well as the observed form for the field dependence of the switching time and maximum current.

The total number of Barkhausen pulses differs quite considerably from crystal to crystal. These differences could reflect perhaps, different imperfection concentrations and it would be of interest to learn more about the nature of these nucleating sites. Also, it is not understood at present why, over a period of hours or days, appreciable changes in the number of pulses from a given crystal sometimes occur. Certainly more investigation into how the pulse count depends on the immediate past history of a crystal is indicated.

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APPENDIX. FORMULATION OF THE CURRENT WAVE FORMS

It is proposed to show that the model of the switching sequence suggested by the Barkhausen pulse studies yields current wave forms in reasonable agreement with those observed experimentally. In particular it will be supposed that spike-shaped nuclei extending more or less through the crystal thickness appear randomly according to the nucleation law and then proceed to expand sideways in all directions. Also, it will be assumed that this model still applies at field strengths sufficient to cause switching to be completed within a few microseconds. At these speeds, slow time effects caused by relaxing space-charge fields will be unimportant. The mathematical approach is standard in nucleation theories.[19]

For random nucleation, the nucleation rate is

$$dN/dt = k_1(N_0 - N), \tag{5}$$

where N is the number of nucleations that have occurred and k_1 is the nucleation probability, that is,

$$k_1 = g\exp(-\alpha/E), \tag{6}$$

where g is a frequency factor. Thus,

$$(dN/dt)_{t=t'} = k_1 N_0 e^{-k_1 t'}. \tag{7}$$

The growth of a nucleus by sideways expansion may be represented by the expansion of the area that it subtends at the crystal surface, namely, s. It is now necessary to choose particular models for the time variation of s. We note that the nucleus is regarded as expanding radially and it seems a reasonable assumption that the wall velocity, c (that is, the radial velocity), is roughly proportional to the field. Thus,

$$c = kE, \tag{8}$$

where k is a constant. Equation (8) tacitly assumes that E is much greater than a threshold field, if the latter exists.[20] Therefore, the area, s, of a nucleus that appeared at time t' increases such that

$$s_{t-t'} = c^2(t-t')^2. \tag{9}$$

The total area covered by all those nuclei that have appeared up to time t is then given by

$$\begin{aligned} S(t) &= \int_0^t s_{t-t'}\left(\frac{dN}{dt}\right)_{t=t'} dt' \\ &= k_1 c^2 N_0 \int_0^t (t-t')^2 e^{-k_1 t'} dt' \\ &= \frac{2c^2 N_0}{k_1^2}\left[\tfrac{1}{2}k_1^2 t^2 - k_1 t + 1 - e^{-k_1 t}\right]. \end{aligned} \tag{10}$$

Now the switching current i varies as dS/dt. Thus

$$i(t) \propto \frac{2c^2 N_0}{k_1}\left[k_1 t - 1 + e^{-k_1 t}\right]. \tag{11}$$

When t is small, i varies as t^2. At low fields, that is, less

[19] P. W. M. Jacobs and F. C. Tompkins in *Chemistry of the Solid State*, edited by W. E. Garner (Academic Press, Inc., New York, 1955), Chap. 7.

[20] H. H. Wieder, J. Appl. Phys. **27**, 413 (1956).

than 5×10^3 volts cm^{-1}, the switching transient does indeed show an initial increase of i with t that is faster than linear. It is interesting to note that this faster-than-linear dependence of the total current results from the above types of calculation only if the area of each individual nucleus expands more rapidly than linearly with t. In particular, wedge-shaped nuclei expanding by sideways motion of their major faces (that is, linear rather than radial expansion) yield a current transient for which $d^2i/dt^2<0$ at small t.

Equation (11) was obtained assuming that the domains expanded indefinitely. If it is supposed that they expand up to an average size S_1 then (11) is no longer valid after the first nuclei have reached the size S_1. This takes a time $t_1=S_1^{\frac{1}{2}}c$. For times $t_2>t_1$, we now have

$$S(t_2)=\int_{t_2-t_1}^{t_2} S_{t_2-t'}\left(\frac{dN}{dt}\right)_{t=t'}dt' +S_1\int_0^{t_2-t_1}\left(\frac{dN}{dt}\right)_{t=t'}dt'$$

$$=k_1c^2N_0\int_{t_2-t_1}^{t_2}(t_2-t')^2e^{-k_1t'}dt' +S_1k_1N_0\int_0^{t_2-t_1}e^{-k_1t'}dt'$$

$$=\frac{c^2N_0}{k_1^2}[-2e^{-k_1t_2}+e^{-k_1(t_2-t_1)}(k_1^2t_1^2-2k_1t_1+2)] -N_0S_1[1-e^{-k_1(t_2-t_1)}]. \quad (12)$$

Differentiating with respect to t_2 gives the current variation, namely,

$$i(t_2)_{t_2>t_1}\propto A\exp(-k_1t_2), \quad (13)$$

where A is a constant given by

$$A=[(c^2N_0/k_1)(2-ak_1^2t_1^2+2ak_1t_1-2a)-N_0S_1k_1a],$$

and $a=\exp(k_1t_1)$. Thus the current transient predicted by this model has an exponential tail of decay constant k_1^{-1}.

The current transient predicted by the above model is compared with an actual transient obtained at a field of about 4000 volts cm^{-1} in Fig. 17. The decay of the pulse was found to be very close to an exponential and from the semilogarithmic plot a value for k_1 was obtained. Substituting this value into the expression in brackets in Eq. (11) allowed the form of the initial growth of the current to be calculated. In the figure the ordinate of the theoretical curve for small t was scaled so as to intersect the straight line at the time corresponding to the maximum of the current transient. It is seen that the agreement over the rising part of the transient is not very good though it is felt to be

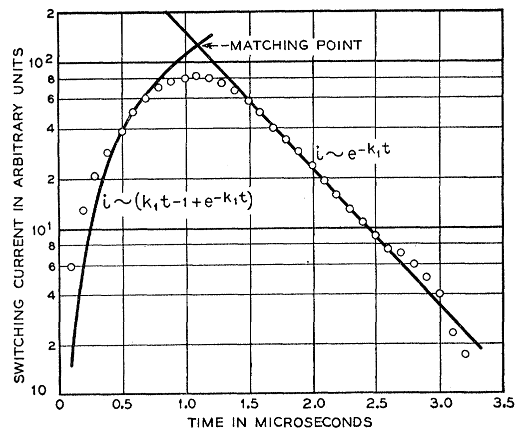

FIG. 17. Comparison between the observed switching transient and the theoretical curves based on a nucleation model applicable at low fields.

reasonable in view of the assumptions that have to be made regarding the mode of expansion of the nuclei.

It is of interest to predict the field dependence of the switching time, t_s, and the maximum current on the basis of the above model. The switching time is defined as the time taken for all but a small fraction of the crystal to switch. Thus, if the total crystal area is S_0, we have $S(t_s)/S_0=\text{constant}$, G. From Eq. (12) it follows that

$$e^{-k_1t_s}=\frac{(k_1/c)^2[(GS_0/N_0)+S_1]}{-2+e^{k_1t_1}[k_1^2t_1^2-2k_1t_1+2+S_1(k_1/c)^2]}. \quad (14)$$

Now G is slightly less than unity while $S_0/N_0=S_1$. Also, at the fields used, c is probably of the order $10^{-2\pm1}$ cm sec^{-1}, while reasonable values for k_1, t_1, and S_1 are 10^5 sec^{-1}, 10^{-6} sec, and 10^{-6} cm^2, respectively. Thus, to a good approximation, (14) reduces to

$$e^{k_1t_s}\propto 2e^{-k_1t_1}, \quad (15)$$

and in particular, t_s varies linearly with t_1 which in turn, varies inversely as the sideways wall velocity. Thus $1/t_s$ varies linearly with the field, in agreement with experimental observations.[1]

The maximum current will occur when $t\approx t_1$ and, therefore, while k_1t_1 is appreciably less than unity. Thus, Eq. (11) can be approximated as

$$i_M\propto\frac{2c^2N_0}{k_1}\cdot\frac{k_1^2S_1}{2c^2}.$$

Thus, i_M varies as k_1, or

$$\ln i_M\propto-(\alpha/E), \quad (16)$$

which, again, is the observed behavior.[1]

At fields higher than those supposed to be present for the above discussion, the expanding domains may be able to surmount the potential barriers between different nucleating regions and so overrun some adjacent sites before they have had a chance to nucleate. These continually expanding nuclei will eventually cease to grow when they encounter other growing nuclei. Thus, the mathematics has to take into account the amount of overlap of domains and the number of nucleating sites that do not get a chance to nucleate. This problem has been solved for the case of random nucleation.[19] We have

$$-\ln[1-F(t)]=\frac{2c^2k_1N_0}{S_0}[\tfrac{1}{2}k_1^2t^2-k_1t+1-e^{-k_1t}], \quad (17)$$

where $F(t)$ is the fraction of the crystal switched at time t. The switching current, obtained by differentiating with respect to t, is

$$i(t)\propto\frac{2c^2k_1^2N_0}{S_0}[k_1t-1+e^{-k_1t}]\exp\left(-\frac{2c^2k_1N_0}{S_0}\times\left[\frac{k_1^2t^2}{2}-k_1t+1-e^{-k_1t}\right]\right). \quad (18)$$

When t is large, e^{-k_1t} is small and the current can be expressed, approximately, as

$$i(t)\propto\frac{2c^2k_1^3N_0}{S_0}t\exp\left(\frac{c^2k_1^3N_0}{S_0}t^2\right),$$

or

$$i(t)=\lambda t\exp(-\mu t^2), \quad (19)$$

where λ and μ are constants. It is interesting to note that this is the same form that has been found, empirically, to fit the switching transients at high fields.[21] [Alternatively, if k_1t is large, the transient takes on the form

$$i(t)=\lambda' t^2\exp(-\mu' t^3).$$

It is well known that the shape of the transient can be modified appreciably by the nature of the voltage cycling the crystal receives.]

Again it is possible to make predictions as to the field dependence of the switching time and maximum current. At $F(t_s)=G$, we can write

$$\ln(1-G)\simeq c^2k_1^3N_0t_s^2/S_0,$$

or

$$c^2k_1^3t_s^2=\text{const.}$$

Hence t_s varies as $(c^2k_1^3)^{-\frac{1}{2}}$ and, from Eqs. (6) and (8), t_s varies as

$$(kE)^{-1}[1+(3\alpha/2E)+\cdots].$$

Therefore, t_s varies as E^{-1} if E is sufficiently large compared with α. This also, is in line with experiment.[1]

Finally, assuming that Eq. (19) is a reasonable approximation at t_M, the time at which the current is a maximum, we find, by putting $(di/dt)=0$, that

$$t_M=(S_0/2N_0c^2k_1^3)^{\frac{1}{2}}. \quad (20)$$

Substituting this in (19) yields

$$i_M\sim ck_1^{\frac{1}{2}}=kE[1-(\alpha/2E)+\cdots]. \quad (21)$$

Thus, in this case, the maximum current varies linearly with the applied field if E is sufficiently large with respect to α. This again is in keeping with experiment.[1]

Note added in proof.—Since this paper was written, Robert C. Miller of these Laboratories has obtained direct evidence that Barkhausen pulses of similar form to those described in this paper can be generated when two domain walls in sideways motion approach each other closely. Thus, there is some doubt as to whether Barkhausen pulses are generated when a new domain is nucleated. However, there is still no evidence that Barkhausen pulses represent jerky wall motion of the sort discussed in this paper, that is, where the magnitude of the pulse is determined by the inclusion size or the distance between inclusions. Miller has also invoked the depolarizing fields to account for the Barkhausen pulses generated by the close approach of two domain walls (to be published). Whether or not Barkhausen pulses represent domain nucleations, the formulations of the switching transients still hold in the framework of the assumptions made in deriving them, namely, a definite number of nucleating sites giving rise to random nucleation of domains followed by sideways expansion.

[21] C. F. Pulvari and G. E. McDuffie, Communications and Electronics, Am. Inst. Elec. Engr. **28**, 681 (1957).

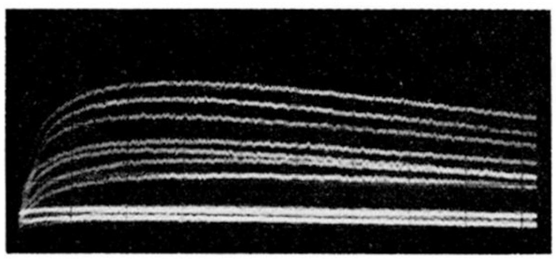

FIG. 1. Oscilloscope photographs of typical Barkhausen pulses. The total length of the trace is 100 μsec.

FIG. 15. Oscilloscope photograph showing some of the abnormal pulse shapes that were occasionally observed.