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**Applied
Electromagnetics
in
Materials**

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in

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*Proceedings of the First International Symposium
Tokyo, 3-5 October 1988*

Edited by

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Preface

The International Symposium on "Applied Electromagnetics in Materials" was held in Tokyo, Japan, on 3-5 October 1988 under the sponsorship of the Ministry of Education, Science and Culture and the organization of an international steering committee.

The presence of this symposium in the scope of applied electromagnetics reflects closely the recent significant progress achieved, in particular, in numerical simulations of electromagnetic field analysis and in superconducting technology and its applications after the last two successful IUTAM symposia, the IUTAM-IUPAP Symposium on "Electromagnetomechanical Behavior of Solid Continua" held in Paris in July 1983, and the IUTAM Symposium on "Electromagnetomechanical Interactions in Deformable Solids and Structures" held in Tokyo in October 1986.

It is believed through recent tendencies observed in the field that various kinds of applications of electromagnetic phenomena could be understood within the area of applied electromagnetics. The scope of this symposium is thus focused on applied electromagnetics in materials, and attempts to provide an international forum to bring together a group of researchers in the field and to exchange innovative ideas and information in the following specified fields:

1. Electromagnetosolid Mechanics
2. Applied Electromagnetics in Free Electron Lasers
3. Application of Electromagnetic Force and Phenomena
4. New Approaches in Eddy Current Analysis
5. Micromechanics in Electromagnetic Fields
6. Applied Electromagnetics in Superconducting Materials

A total of 70 participants represented a wide spectrum of industrial, research institutes and universities from Japan as well as other countries. To highlight researches in the fields, the program committee initiated the six sessions by invited papers from distinguished scholars currently working in the fields. 32 contributed papers were also presented orally at the symposium. The authors were requested to prepare their papers in "camera ready" form which are now included in these proceedings. We are appreciative of the cooperation of all the contributors.

Finally, it is a pleasure to express our appreciation to the local organization committee from the staffs, Professor T. Takagi, Mr I. Saito, Mr Hashimoto and Mr G. Yoshizawa, of the Nuclear Engineering Research Laboratory of the University of Tokyo for their preparation assistance to the success of this symposium.

December 1988

Kenzo Miya
Richard K.T. Hsieh

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I.

Electromagnetosolid Mechanics

Coupled Magnetomechanical and Electromechanical Hysteresis Effects

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ABSTRACT

After a brief review of mechanisms at the basis of microscopic and macroscopic electric (or magnetic) hysteresis, a thermomechanical phenomenological approach is presented which allows one first to reproduce electric (or magnetic) hysteresis loops and, second, to account for the reciprocal coupling between this hysteresis and mechanical fields (applied stresses, induced strains). These couplings can be exploited in non-destructive testing techniques.

KEYWORDS

Hysteresis ; electroelasticity ; magnetoelasticity ; applications.

INTRODUCTION

The reciprocal coupling between magnetic or electric hysteresis, respectively in hard ferromagnets and ferroelectric ceramics, and stresses is examined in the light of continuum thermodynamics, and the consequences of these effects in nondestructive testing and material science studies are discussed. First, it must be noticed that ideal hysteresis loops and real, observed ones in magnetic and electric materials differ in essential ways. Ideal (square-type) loops pertain to monocrystals and monodomain samples. Real, rounded off, hysteresis loops present a *hardening* which can be reproduced only in a phenomenological theory for multidomain bodies. This magnetic or electric "hardening" can be related to the Barkhausen effect and the characteristically stress-dependent features of the latter can be used as an NDT means of studying stresses by a magnetic or electric method. Real hysteresis loops have a final shape which captures in a single picture a lot of information (for instance, the influences of temperature, bias fields, history of loading, stresses, irradiation, ...). On the one hand, condensed-matter physics avoids hysteresis phenomena by introducing the notions of phase transition, Maxwell rule, and convexification of the free energy function. In more mathematical terms, it avoids the cusp catastrophe. This is briefly discussed. The main point, however, is that no

dissipative processes are involved in this description, and this quite contrarily to experimental evidence. In contrast, the approach presented acknowledges the presence of hysteresis and its complexity, and puts it in a thermodynamical framework which fully accounts for dissipation and the various, above-mentioned influences. The phenomenological theory thus addressed to, like other modern theories of dissipative behaviors (plasticity, viscoplasticity), uses the notion of *internal* state variable. While several general theorems related to stability (magnetic or electric analogues of the principle of maximal dissipation of Hill, of Drucker's inequality, of Ilyushin stability postulate) can be proved, it is also reminded that a microscopic support can be given to this phenomenological presentation in terms of domain wall motions. The influence of bias stresses on hysteresis loops (changes in saturation, coercive field, and instantaneous susceptibility) is established in agreement with experimental data. Simultaneously, the same general framework can be used to construct a theory of poling and spontaneous induced strains in ferroelectric ceramics. Pure theory thus joins physical data and experimental methods in an harmonious whole.

NOTION OF HYSTERESIS

Ideal and observed hysteresis loops

It is wellknown that the characteristic feature of a *ferroelectric* behaviour is the appearance of a spontaneous electric dipole moment which can be reversed, without change in magnitude, by an applied electric field. This occurs in the *ferroelectric* phase within a certain range of temperature, a *phase transition* existing at the boundaries of this range. Dielectric anomalies (yielding, in turn, anomalous elastic and calorimetric behaviours) are exhibited at this phase transition. Here we focus on the electric hysteresis. On varying the voltage V across the plates of the parallel plate condenser in Fig. 1(a) one obtains the typical "square" response ABCDE'C'D' in Fig. 1(b). When the voltage lies between values

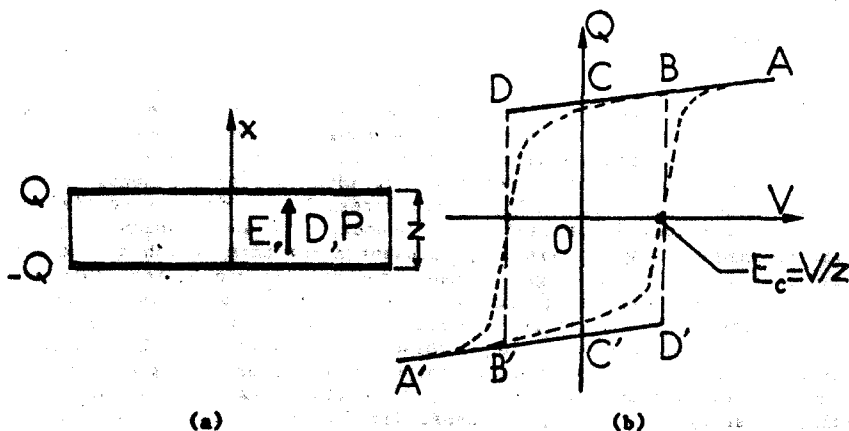


Fig. 1 Ideal and observed hysteresis loops from the parallel plate condenser. The voltage V induces charges Q and $-Q$ on the opposite plates.

at D and D' , then there are two possibilities and the electric charge on a given plate is determined by the *past history* of the system, i.e., whether we are electrically loading or unloading the specimen. In effect, the *sign* of the last applied voltage lying outside the switchover values determines the branch describing the response in electric charge. This is the essence of *dielectric hysteresis* as of all hysteretic phenomena for that matter. For electric processes this is usually represented on a diagram giving electric displacement $D = Q/a$ (or electric polarization P) versus electric field $E = V/z$. This "scaling" provides the constitutive equation $E(D)$. The value of the electric field required to cause a switch from one branch to the other is called the *coercive field*. The electric response thus described is ideal. Observed hysteresis loops are more or less tilted on the vertical axis and they are rounded, looking like the dashed hysteresis curve in Fig. 2(b). The reason for these "imperfections" is the occurrence of a structure in domains and walls (which is energetically more favourable) in the slab of Figure 1(a). These domains are small regions with electric state equation presumably of the ideal type pictured in Fig. 1(b) (they are nearly saturated with spatially uniform properties). The electric dipole moments of adjacent domains are not parallel so that the electric field and displacement are *not* uniform in the slab. This results in a rounding off of the ideal hysteresis loop which, therefore, corresponds to a monodomain. On this real hysteresis loop the coercive field is some average electric field in the crystal. It is connected to the single-crystal properties in some complicated manner. The real hysteresis loops are obtained in successive phases with complete or partial rearrangements of domains, the steepest parts of the hysteresis loop corresponding to a rapid switching of many domains, the tails to a tendency to reaching a single-domain state, and those parts nearby zero electric field to the fact that only very few of the oriented domains return to their initial state on switching off the electric field. Investigations of the dependence of the polarization on the field, carried out using sensitive equipment, show that an increase in the polarization is frequently *discontinuous* (Fig. 2), suggesting that the polarization reversal occurs suddenly in small parts of a crystal. The reversal of polarization in such a small region, a fraction of a domain,

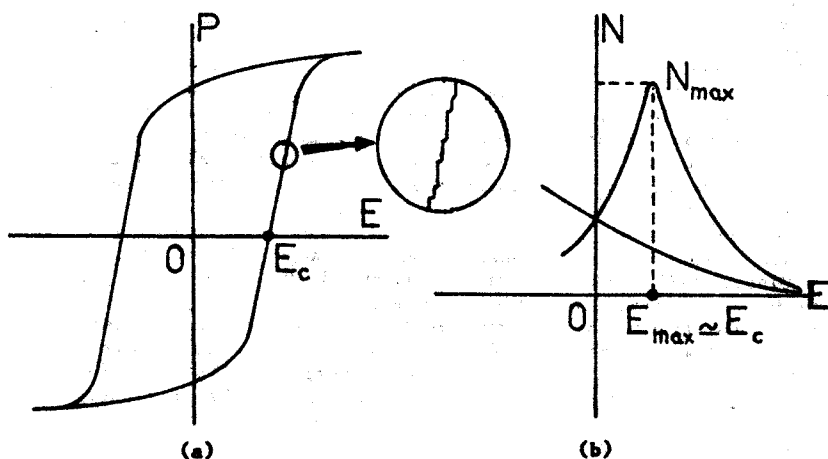


Fig. 2 Barkhausen jumps : (a) hysteresis loop ;
(b) recording.

is called a *Barkhausen jump*. The resulting "rapid" polarization reversal followed by subsequent "immobility" can be explained by the presence of various structural defects (growth defects, dislocations, impurities) which impede the motion of a domain wall. This is observed as a noise called the *Barkhausen noise*. The recording amounts to counting Barkhausen jumps and this is strongly correlated to the slope of the hysteresis loop, the largest number being observed for a value of electric field practically that of the coercive field (steepest part of the hysteresis loop). The recording looks like an upside down butterfly for a regular symmetric hysteresis loop (Fig. 3).

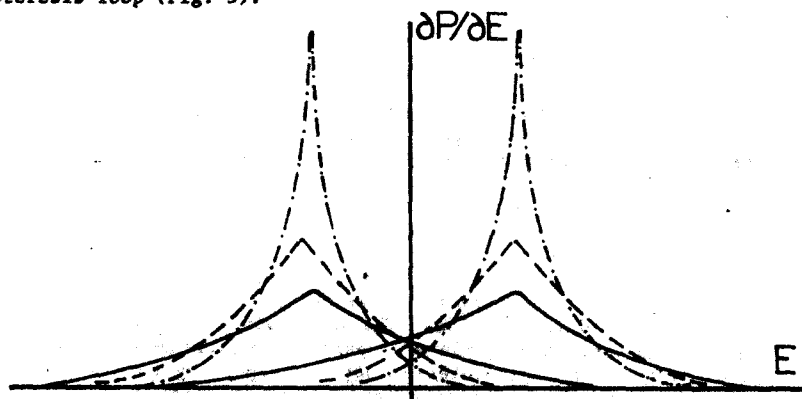


Fig. 3 Instantaneous electric susceptibility vs. electric field (note resemblance with Fig. 2(b)) for various applied stresses.

Energy-based approach

The dielectric loop is a function of various parameters such as temperature, stresses, irradiation, *etc.*, the first of these being the essential parameter in relation to phase-transition theory. The phase-transition viewpoint in the manner of Landau, Ginsburg (1946) and Devonshire (1954) refers only to *nondissipative* processes, the discussion being entirely based on the expression of the free energy and the stability of various parts of the electric response curve. This is a simple way of looking at multivalued responses which may eventually produce hysteretic effects. In this type of approach one essentially looks for an expansion of the free energy in terms of the most characteristic parameter (the order parameter, e.g., P) and then examine the stability of the response depending on the sign of coefficients in the expansion and the range of temperature (cf. Lines and Glass, 1977, Chap. 3, and Grindlay, 1970, Chap. 5). Typically, one obtains a representation such as in Fig. 4 with the appearance of second-order phase transition and hysteresis with a decrease in temperature (t = reduced temperature, G = Gibbs energy). *Second order phase transition* corresponds to considering the schematic response $A'B'EA$ in (a) following Maxwell's construction and the convexification of the free energy density. The hysteresis loop is considered if the *metastable* parts CB and $B'C'$ are allowed. But this is very schematic.

Like mechanical hysteresis, the electric hysteresis of real industrial electric materials (polycrystals, multidomain structures) is a complex phenomenon and there is need, at the moment, for a phenomenological approach

relying on irreversible thermodynamics and providing the best possible reproduction of real hysteresis curves.

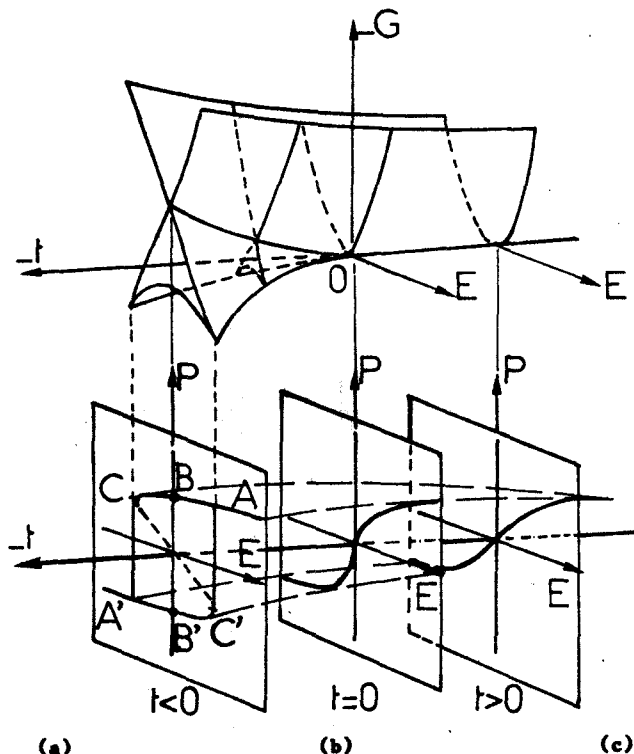


Fig. 4 Unfolding of the cusp catastrophe and appearance of second-order phase transition or hysteresis in ferroelectrics

Real hysteresis loops

Barium titanate (BaTiO_3) is a prototype ferroelectric single crystal of the displacive type, which presents a definite saturation of the polarization due to domain reorientation, and whose hysteresis loops are nearly rectilinear with a coercive field varying from 500 to 2000 volt/cm. Triglycine sulfate (TGS) exhibits hysteresis loops which are very nearly perfect rectangles. In a very narrow range of electric fields close to the coercive value, almost all domains reverse their polarization and the induced polarization is practically negligible compared to the spontaneous moment. At room temperature the coercive field is about 400 V/cm in an applied polarizing field of 1500 V/cm amplitude at 50 cps. However, such regular and symmetric loops can be drastically distorted by an external agent such as irradiation by gamma rays. For instance, Fig. 5(b) represents the shifted and distorted loop of TGS irradiated by gamma rays. The latter cause both a displacement, which can also be obtained by applying a bias electric field, (see Zheludev, 1971, vol. 2, p. 426), and a distortion marked by the appearance of multiple inflexion points. This is more visible

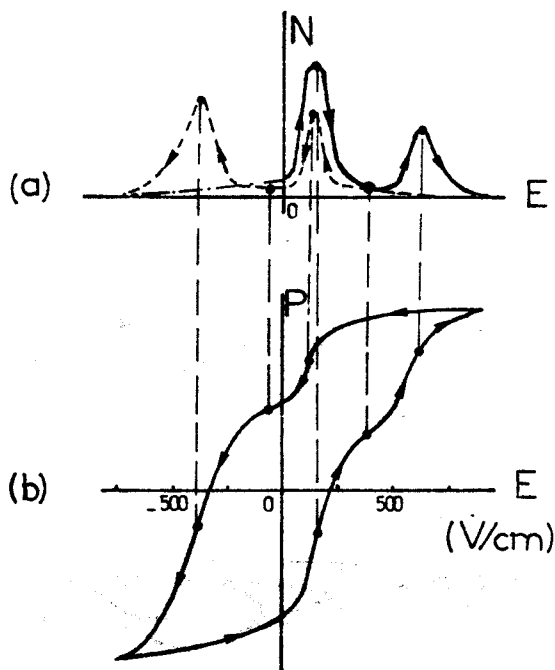


Fig. 5 (a) Distribution of Barkhausen jumps vs electric field and (b) distorted hysteresis loop for γ -ray irradiated triglycine sulfate.

on the "derivative" curve 5(a) obtained experimentally (Rudyak, 1971, p. 476), which is none other than the distribution of Barkhausen jumps with two maxima on loading and two maxima on unloading, these being placed nonsymmetrically with respect to the vertical axis.

Polycrystalline ceramics such as tetragonal PLZT 6.25/50/50 ceramics exhibit a rather smooth hysteresis loop at room temperature with a high coercive field in low frequency electric loading.

The coercive field of a ferroelectric depends on the temperature. A general feature of this dependence is that the coercive field increases when the temperature is reduced from the Curie phase-transition temperature. The increase is due to a decrease in the mobility of domain walls.

Stresses also influence the shape of electric hysteresis loops. for instance, the loop of PZT ceramics is markedly sensitive to the action of stress as evidenced by Fig. 6 which exhibits the influence of a stress field applied parallel to the direction of the electric field and electric polarization. The maximum (saturation) and remanent polarization as well as the coercive field clearly decrease with increasing compressive stress while for a stress perpendicular to the field the influence is markedly small but the coercive field slightly increases with increasing compressive stress. There is thus globally a marked coupling between the ferroelectric hysteresis loop and the state of stresses, that the latter be reversible

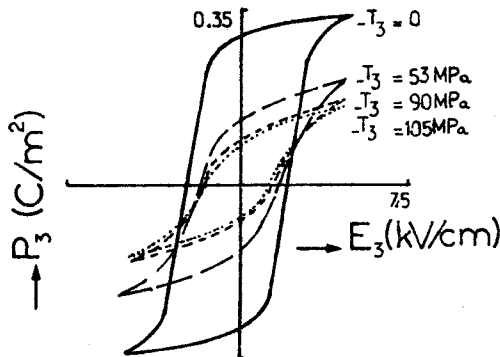


Fig. 6 Influence of stress on dielectric hysteresis loop of PLZT ceramics (after Arndt *et al.*, 1984) : loop $P_3(E_3)$ for different compressive stresses $-T_3$.

or residual. These couplings are certainly *nonlinear*, as shown by the influence of compressive stress on saturation level in Fig. 6.

THE PHENOMENOLOGICAL PROBLEM

In brief, we consider the following problem. A polycrystalline, multidomain dielectric material exhibits both *induced* electric polarization and *spontaneous* electric polarization. The former is reversible while the latter is accompanied by dissipation. In the reference configuration of the material these two polarizations are noted \mathbf{H}^I and \mathbf{H}^R , respectively, the total polarization being none other than the vectorial sum of these two :

$$\mathbf{H} = \mathbf{H}^I + \mathbf{H}^R. \quad (1)$$

We focus on \mathbf{H}^R and its relation to the polarizing field and other influences such as temperature, stresses, and bias electric fields. For a given temperature and zero stress and bias field, the response $\mathbf{H}^R(\mathbf{E})$ presents the shape of a *hysteresis* loop (a) in Fig. 7(a) with saturation such that

$$|\mathbf{H}^R| \rightarrow \mathbf{H}_S^R \quad (2)$$

as $|\mathbf{E}|$ goes to infinity. This hysteresis loop is obtained in an alternating electric field of *low* frequency. It is *assumed* that the hysteresis loop thus obtained does *not* depend on that frequency. In other words, the electric hysteresis phenomenon here is considered to be *rate-independent* in the first approximation (i.e., it does not depend on the time rate $\dot{\mathbf{E}}$ of the polarizing field) and, as such, it does not involve any characteristic time in contradistinction, say, with viscosity or electric relaxation. This is probably not true for high frequencies of the polarizing field and for relatively high temperatures. The hysteresis curve (a) in Fig. 7 is essentially characterized by (i) the level of saturation, (ii) the value of the coercive field $E_c = E(\mathbf{H}^R=0)$, and (iii) the inclination of the hysteresis loop on the \mathbf{H}^R -axis (remember that perfect electric hysteresis loops have vertical, jump-like branches at the coercive-field

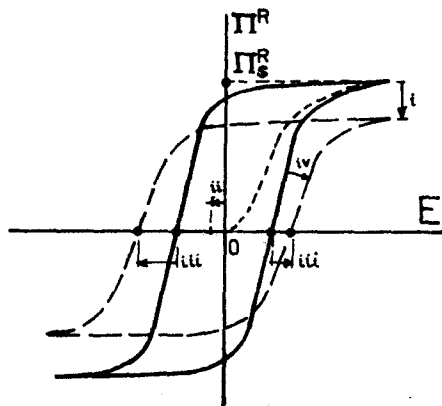


Fig. 7 Typical hysteresis behaviour of ceramics, (a) in the absence of applied stress and bias electric field ; (b) in the presence of such fields.

value ; see Fig. 1(b). When this inclination is not zero, what appears to be the case for most industrial materials such as ceramics, then we say that the electric material exhibits *electric hardening* (i.e., it takes a greater value of the polarizing field to increase its polarization Π^R by a given amount). This wording is granted in total analogy to *mechanical hardening*.

Under the influence of a perturbation such as a bias electric field, an applied stress or irradiation, the loop (a) transforms into loop (b) in Fig. 7. In general, four essential effects are manifested : (i) the saturated level has changed ; (ii) the width of the hysteresis loop at zero polarization has been altered ; (iii) the coercive field has evolved, presenting different values on loading and unloading (the loop is no longer symmetric with respect to the origin), and (iv) the electric hardening has been modified to some extent, all facts which are even more clearly visible on the derivative curve of the hysteresis loop (i.e., the instantaneous electric susceptibility) or, for that matter, on a count of Barkhausen jumps as in Fig. 2(b). The purpose of the following sections is to present a thermodynamically admissible phenomenological theory, of necessity *nonlinear*, which allows one to build the hysteresis loop (a), starting from a virgin state, and to reproduce the above-described effects on that hysteresis loop. This theory is due to Bassiouny *et al* (1988a,b ; 1989) and Maugin and Bassiouny (1988) ; it closely follows a somewhat similar nonlinear theory built previously for magnetomechanical hysteresis (Maugin *et al*, 1987) and which receives the support of the semi-microscopic theory of domain-wall motions in a defective material (Sabir and Maugin, 1988). It has much in common with the elastoplasticity of materials with locking (Suquet, 1985).

IRREVERSIBLE THERMODYNAMICS OF ELECTRIC HYSTERESIS

According to the working hypotheses considered above electric hysteresis is viewed as a dissipative process without time scale, but it accounts for the past history of the electric loading of the sample in some manner. The first point is coped with by assuming that the power dissipated in the time evolution of Π^R is homogeneous of degree *one* in Π^R , while