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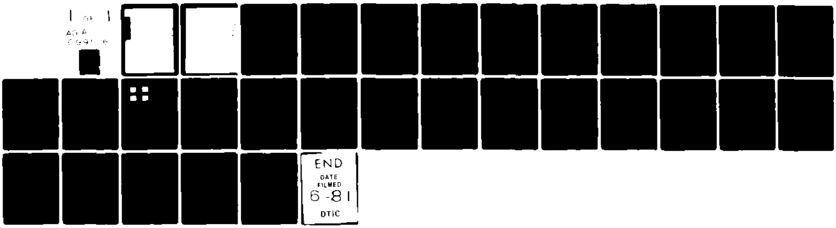

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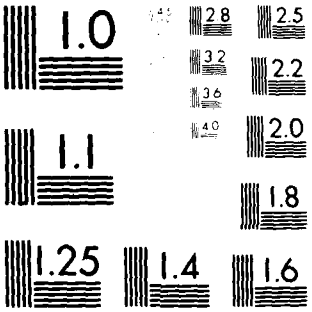

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The feasibility of a new kind of nonvolatile digital memory is proposed and examined. The basis of the memory is an anomalous photovoltaic phenomenon found in ferroelectric ceramics--a photovoltage with a polarity that depends on the direction of the remanent polarization. Described are proposed memory cell structures that function in matrix arrangements as nonvolatile read/write random access memory or electrically alterable program-

20. Abstract (Cont'd)

mable read-only memory. Test results obtained with an experimental test unit also are described. Included is a survey of methods for producing ferroelectric films.

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FIGURES

1. INTRODUCTION

This paper proposes and experimentally examines the feasibility of a new kind of nonvolatile digital electronic memory. The basis of the memory is an anomalous photovoltaic phenomenon found in ferroelectric ceramics--a photovoltage with a polarity that depends on the direction of the remanent polarization. Opposite-polarity photoelectromotive forces (emf's) are associated with opposite directions of remanent polarization, and the magnitude of the photo-emf is linearly proportional to the magnitude of the remanent polarization.*

Various schemes to use this phenomenon for storage and retrieval of information have been described. In all of these schemes, information is stored within an element or a substrate as direction and magnitude of remanent polarization and is retrieved by sensing the polarity or the magnitude of an emf (a photo-emf) produced by the illumination.†

The proposed digital electronic memories combine ferroelectric and semiconductor elements electrically in addressable matrix cells. Binary form digital data are stored as remanent polarization within an array of illuminated ferroelectric elements. The elements are polarized in either of two opposite directions with voltage pulses by using transmission gates. They are polarized in a manner analogous to that in which a gating transistor transfers charge on the gate of a sense transistor in dynamic random access memory (RAM). The element is illuminated (illumination is from a low-intensity steady source), and a photo-emf positive or negative (depending on the direction of polarization in the element) develops across the element. This emf is in series with the gate of a field-effect transistor (FET) (sense transistor) associated with each element. The result is either of two voltage levels on the transistor gate (gate biases). These levels produce in the transistor drain-source channels either an "on" (high-conductivity state) or an "off" (low-conductivity state). Once the direction of remanent polarization within an element is switched (reversed), a particular polarity steady photo-emf results, and the transistor is in one of its two possible conduction states. The transistor remains in that state until the direction is switched again. Removing the illumination at any point results in a temporary loss of the controlling photo-emf. Restoring the illumination restores the original emf and transistor conduction state. Memory is retained in a completely unpowered, unilluminated state.

In this photovoltaic ferroelectric-semiconductor memory, the electrode on a ferroelectric element can be connected with a conductor to the gate metalization of a conventional FET. The approach is distinct

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See Selected Bibliography, Photovoltaic Phenomenon.

†*See Selected Bibliography, Storage and Retrieval.*

from that in which ferroelectrics interface directly with semiconductors[1] with conduction in the semiconductor modulated by a depolarization field from the ceramic. This is the so-called ferroelectric field effect. Ferroelectric field effect devices require no illumination to function. However, it is a characteristic of such devices that the conductivity change in the semiconductor decays with time,[1] limiting storage times to durations as short as several hours. The photovoltaic devices, on the other hand, appear to be truly nonvolatile. The nonvolatility results from an unchanging (with time) relation between the remanent polarization and photo-emf and the fact that, except for a small initial aging, the remanent polarization in ceramics is stable.

Proposed in this paper are memory cell structures that function in matrix arrangements as nonvolatile read/write RAM or electrically alterable programmable read only memory (EAPROM) devices. Test results obtained with an experimental test unit are described. These results demonstrate how the memory functions, provide experimentally determined read/write times, and show that stored information is retained. The intent is to eventually make devices that are integrated onto a silicon substrate, with the required ferroelectric elements produced by processing compatible with silicon technology. For this reason, included in this paper is a survey of the current status of processes for producing ferroelectric films. Also in this paper, the memory characteristic of a presently available silicon nonvolatile memory of the charge storage type is compared with that predicted on the basis of the experimental results for a functionally similar photovoltaic ferroelectric-semiconductor device.

2. MEMORY CELL

The ferroelectric element within each matrix cell is a two-terminal capacitive structure with metal electrodes. In the test device, the elements were planar structures produced by depositing thin film electrodes onto a ceramic substrate (fig. 1). A voltage pulse applied to the electrodes results in a fringing field within the substrate; the field polarizes the region between the electodes in either of two directions. In the presence of illumination, a photo-emf is developed across the electrodes. In the nomenclature of photoconductivity, such a configuration is called transverse. In a longitudinal structure, the remanent polarization is perpendicular to the surface of the substrate and the photo-emf is developed by illumination through a transparent electrode. In principle, such a structure also could be used.

A proposed read/write cell is shown in figure 2. One electrode of the ferroelectric element, F, is connected directly to the gate of an n-channel enhancement insulating gate field-effect transistor (IGFET),

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[1] *D. C. Burtoot and G. W. Taylor, Polar Dielectrics and Their Applications, University of California Press, Los Angeles (1979), 351.*

which is the sense transistor, Q_1. Two parallel back-biased diodes, D_1 and D_2, connect these to a common return. To write into a cell, a positive or negative write data pulse is applied to the input at transmission gate Q_2 (column select) coincident with a write select signal (row select) to the control inputs of the transmission gate. The diode characteristics result in a high impedance for the small photo-emf shifted gate biases, which have values of several volts or less, but a low impedance for the larger amplitude voltage pulses applied with the transmission gate. As a result of this reduced impedance, voltages in excess of the coercive field appear across the ferroelectric elements and switch the direction of the remanent polarization. If the remanent polarization is initially directed away from the gate of Q_1, a negative write pulse switches the direction toward the gate. Since the memory element is illuminated, a spontaneous photocurrent flows, and a positive voltage (photo-emf) appears across the element. The direct current (dc) impedance of the illuminated ferroelectric element is considerably less than the gate to ground impedance so that the element photo-emf causes the gate bias initially set at the threshold for conduction, V_g, to increase in magnitude. The result of this is a low-resistance state drain-source channel in transistor Q_1.

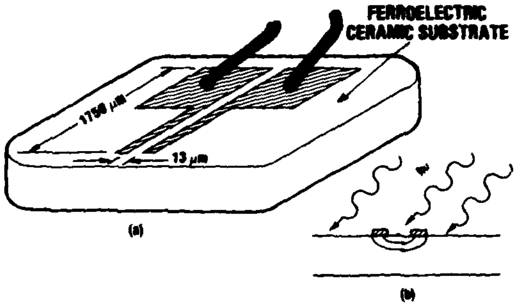

Figure 1. Planar structure ferroelectric memory element as used in test device: (a) view from top and (b) cross section showing fringing field and illumination.

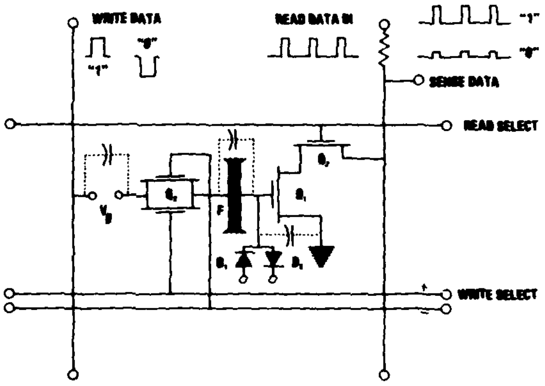

Figure 2. Proposed read/write RAM cell: diodes D_1 and D_2 are back biased; V_g is set at gate threshold for conduction.

Under these conditions, only a small fraction of an input read data pulse (column select) applied through the read select transistor, Q_3, by a read select pulse (row select) appears at the sense output. These low-level signals constitute a logic 0 output. In a similar fashion, a positive pulse writes a logic 1 by switching the element remanent polarization that is initially directed toward the gate of Q_1 so that it is directed away from the gate. Illumination produces a negative photo-emf, which reduces the gate bias to a value below the threshold for conduction in the drain-source channel. The channel is then in a high-resistance state, and a major portion of a read data pulse applied through Q_3 with a read select pulse appears at the sense output. This high-level signal constitutes a logic 1 output. Thus, a positive write data signal writes and states a logic 1; a negative write data signal, a logic 0.

In principle, other similar configurations could accomplish the functions of the configuration shown in figure 2. For example, a p-channel sense transistor could be used with a positive write pulse producing a logic 0 read output. Another possible arrangement would use a junction field-effect transistor (JFET) rather than an IGFET for the sense transistor, Q_1.

Diodes D_1 and D_2 (fig. 1) in principle can be replaced by a single diffused junction or single breakdown diode, which would conduct in the breakdown mode for one of the write polarities. The input diode of the JFET could be used in this manner.

The RAM cell (fig. 2) can be read immediately after write. A charge is generated by a write pulse on the gate of Q_1 by voltage division between the (parasitic) element capacitance and the gate capacitance. The charge is equal approximately to C_gV_{B1} or C_gC_{B2}, where C_g is the gate to ground capacitance and V_{B1} and V_{B2} are the diode biases. This charge decays through the diode back resistances until the voltage at gate equals the photo-emf shifted bias voltage. The gate voltage never decays below the memory bias voltage, provided that an adequate photocurrent flows. The effect is to allow a logic 1 or 0 to be read immediately after write.

This read/write cell uses a transmission gate within each cell. The device can function as an EAPROM device with block programming as well as a read/write RAM. In general, an EAPROM device would not require the same degree of cell isolation as a RAM. A proposed arrangement minimizes the number of transistors in a matrix cell and would function as an EAPROM device (fig. 3). Application of a voltage pulse to the write data line while simultaneously grounding the read/write select results in an induced remanent polarization in an "erased" illuminated ferroelectric element, F. The polarized element produces a photo-emf that biases the gate of the normally on FET to cut off and then results in a high-level logic 1 output. Row select for read is accomplished by grounding the read/write select. The read data pulse input column is the selected column.

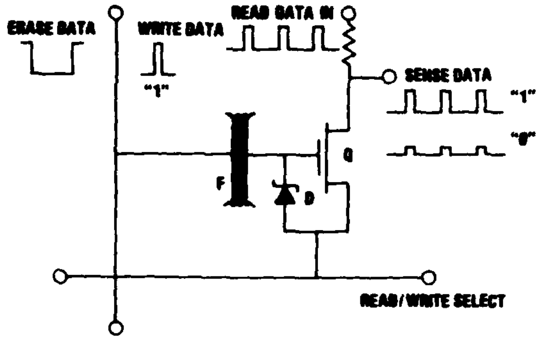

Figure 3. EAPROM cell: write data are applied to "erased" ferroelectric element.

3. TEST DEVICE AND EXPERIMENTAL RESULTS

The test device shown in figure 4 is used to determine characteristics that can be expected from the proposed memory cell shown in figure 2. The write (switching) pulses in the test device are obtained from an instrumentation pulse generator. These could be varied in magnitude, duration, and polarity. The semiconductor components are two low-reverse-current diode pairs and an n-channel enhancement IGFET (sense transistor). The two diodes in series extend the experimental voltage range for switching pulses by increasing the minimum voltage for breakdown. The IGFET is an individually packaged n-channel enhancement device. The ferroelectric element (fig. 1) consisted of metalizations 10 μm wide and 1750 μm long, separated by a 13-μm gap, on the polished surface of a ceramic plate. The ceramic is a polycrystalline solid solution, which is 53 mole percent lead zirconate and 47 mole percent lead titanate, with a 1 weight percent additive of niobium pentoxide. It is usually designated by the acronym PZT-5A.

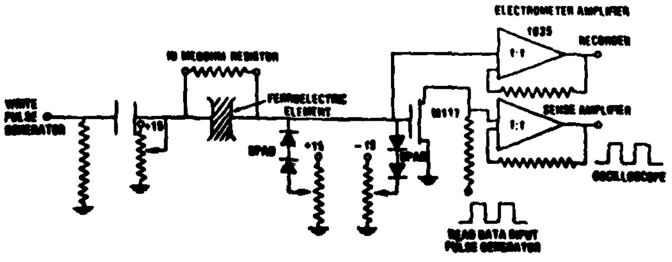

Figure 4. Circuit for test device.

Photovoltages were generated with a uniform flux of broadband ultraviolet illumination distributed about a 325-nm peak. The illumination was produced with a mercury arc filtered successively by a water filter and a CS-7-54 silica glass filter. The illumination could be reduced in intensity over a considerable range by the interposing of fine wire mesh, which served as neutral density filters. The read outputs were displayed on an oscilloscope. A unity gain operational amplifier served as a sense output amplifier. The read data signals were 200-ns, 10-V pulses.

The test device functions in essence as does the proposed cell shown in figure 2. A positive write pulse writes a logic 1, which is read out as high-level (200-ns) output pulses; a negative write pulse writes a logic 0, which is read out as low-level (200-ns) output pulses. The ferroelectric element photo-emf was monitored with an in-situ electrometer amplifier used with a strip chart recorder. The experimental results were obtained with the sense transistor gate biased at its threshold--about 3.5 V. Diode biases V_{B1} and V_{B2} were respectively -6 and +6 V.

In figure 5, the element photo-emf is shown as a function of the number of successive 11-μs, 140-V pulses of a particular polarity applied to the test circuit. The photovoltages were measured about 10 min after the application of a pulse. Each pulse altered the element remanent polarization.

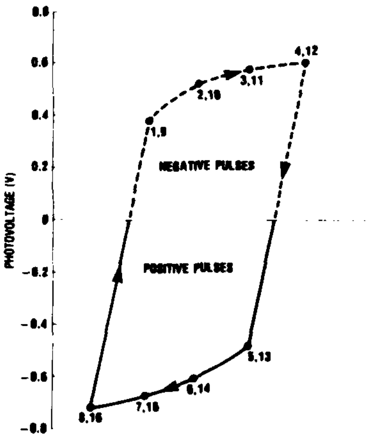

Figure 5. Photoelectromotive forces produced by successive 11-μs, 140-V pulses with polarity switched after 4th, 8th, 12th, and 16th pulses.

The initial state was saturation state produced with several hundred positive pulses. This saturation resulted in a photovoltage of -1 V. A first negative pulse switched the direction of remanent polarization, resulting in a photo-emf across the element of 0.38 V. This is the point indicated by the numeral 1 in figure 5. Additional successive negative pulses (pulses indicated by 2, 3, and 4 in fig. 5) increased the element photo-emf until a photo-emf of 0.6 V appeared across the illuminated element. A fifth pulse of reverse polarity (positive pulse) then switched the direction of the remanent polarization, resulting in a negative photo-emf indicated by the number 5 in figure 5. Additional positive pulses were applied. Each time, the magnitude of the negative photo-emf increased. After the seventh (positive) pulse, the pulse polarity was reversed again, resulting in a positive photo-emf, the same as that produced by the first pulse, as is indicated in figure 5. Additional negative pulses resulted in increased photo-emf's until a 13th pulse (of positive polarity) again reversed the remanent polarization and photo-emf; additional positive pulses were produced and photo-emf's were measured until a final test pulse, pulse 16. The photo-emf resulting from pulse 16 was the same as that resulting from pulse 8.

The results of the two cycles coincided. The results show that for the 11-μs, 140-V pulse, a single switching pulse reverses the polarity of the photo-emf irrespective of the number of pulses of the opposite polarity that preceded it. With the gate bias set at the FET threshold, a combination of element and transistor would function thus in effect (assuming an abrupt turn on voltage) in a saturation mode.

Shorter pulses change the photo-emf, although then a single pulse cannot quite reverse the polarity of the element photo-emf. The results in figure 6 are for 150-ns, 140-V pulses and the same ferroelectric element. There are 70 pulses in each half cycle. The implication of these results is that short pulses could be used to program a block erasable device of the type shown in figure 3, but would not be as suitable as for a true RAM.

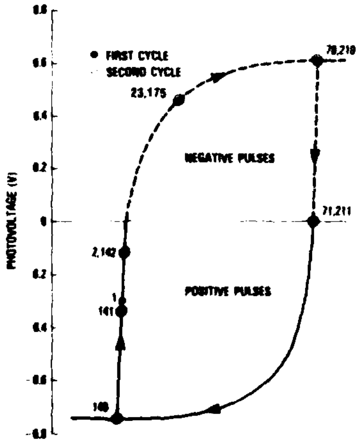

Figure 6. Photoelectromotive forces produced by successive 150-ns, 140-V pulses.

The element photo-emf was measured as a function of illumination intensity for a given remanent polarization state (fig. 7). The photo-emf saturated at about 1 mW/cm^2. Short-circuit current was measured as a function of illuminated intensity by measuring the voltage developed across a shunting resistor (10^8 Ω) placed across the ferroelectric element (fig. 8).

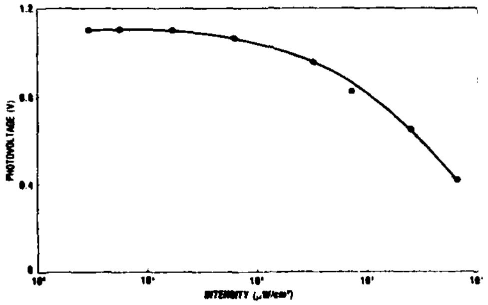

Figure 7. Photoelectromotive forces as function of illumination intensity.

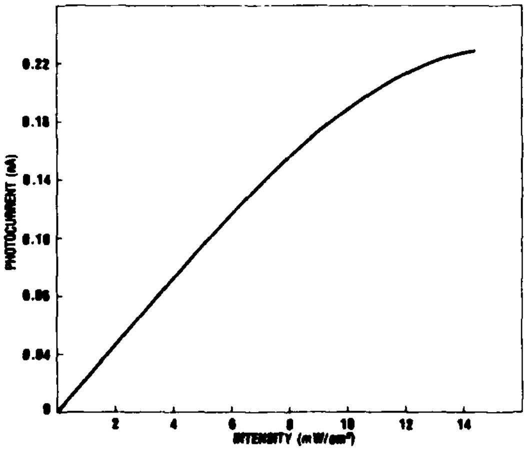

Figure 8. Short-circuit current (zero appplied field) as function of illumination intensity.

The repeated rapid switching of a ferroelectric element could be sustained without detrimentally affecting the memory characteristic. In an experiment, an element was subjected to a series of alternating polarity, 200-ns pulses. The alternating polarity pulses immediately following each other formed pulse pairs, with a 500-kHz repetition rate. This pulse train could be applied for hours (1.8×10^9 switchings/hr) without affecting the element's memory characteristics. The element photo-emf after terminating a pulse train depended only on the polarity of the final pulse. Thus, no fatigue factor was observed after many storage cycles. The current required for switching was measured with a current probe. The value was approximately 4 mA.

Decay of the element photo-emf's (gate bias shifts) with time is shown in figure 9. The write pulses were 600 ns and 140 V. The decay of the photo-emf is initially logarithmic with time. The photo-emf decreases with increasing time until a constant value is reached. This decay characteristic is similar to that of the electromechanical coupling coefficient,[2] which is (as is the photo-emf) proportional to the remanent polarization. Therefore, the decay of photo-emf can be attributed to the aging characteristic of the magnitude of remanent polarization in a ceramic.

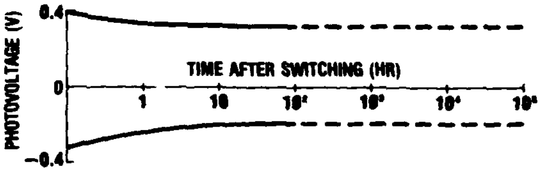

Figure 9. Time history of element photoelectromotive forces: at 100 hr, photoelectromotive force was reversed with switching pulse.

The read output levels are essentially the same at 0.5 and 100 hr. The read outputs are shown in figure 10. The positive bias at the gate photo-emf produced a logic 0; the negative bias at the gate photo-emf produced a logic 1. The time dependencies were obtained under conditions of intermittent illumination. Results under conditions of continuous illumination were similar. It was observed also that the absence or the presence of illumination on a ferroelectric element during switching did not affect in any significant way the pulse voltages or the duration needed for switching.

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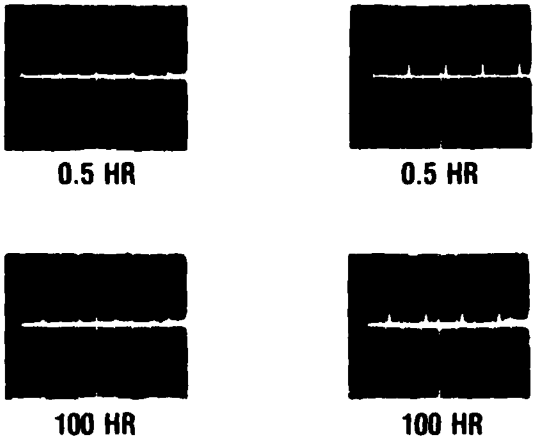
[2] *B. Jaffe, W. R. Cook, Jr., and H. Jaffe, Piezoelectric Ceramics, Academic Press, New York (1971), 83.*


0.5 HR 0.5 HR

100 HR 100 HR

Figure 10. Logic 1 and logic 0 at 0.5 and 100 hr: scale is 2 µs and 5 V per division.

These results show the essential feasibility of electronic memory based on the anomalous photovoltaic effect in the ceramics. The voltages required to write can be reduced by reducing the planar electrode spacing to increase the switching field. Photo-emf's produced with a given electrode spacing can be increased by using finer grain ceramics.[3] Also, reduced coercive fields for switching and increased photo-emf are found in PLZT ceramics. Material of 7/65/35 PLZT has a coercive field of about 14 kV/cm as compared with about 30 kV/cm for the PZT-5A used for the test element. The saturation photo-emf per unit length for 7/65/35 PLZT material of 2- to 4-µm grain size is somewhat larger than that of the PZT-5A producing 1500 V/cm rather than 600 V/cm characteristic of the PZT-5A material used.

4. FERROELECTRIC CERAMIC FILMS

In the test ferroelectric memory element, the photocurrent is generated in a strongly absorbing region within several micrometers of the surface if, as in the test of section 3, the illumination wavelength is shorter than that of the absorption edge. Consider also that polarization reversal is by way of the fringing field. The electrode separation is only 13 µm; as a result, the depth below the surface within which the polarization switches is only several micrometers. The active region of the ferroelectric is thus only a few micrometers thick. Therefore, it seems reasonable that a functional memory element with planar electrodes as in the test device could be fabricated as a film on a supporting substrate. The requirements for such a film are that it be polycrystalline, insulating, and characterized by a remanent polarization and coercive field similar to that of bulk material. Such a film could be expected to produce the anomalous photovoltages characteristic of the bulk ceramic material.

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[3] *Philip S. Brody and Frank Crowne, J. Electron. Mater., 4 (1975), 955.*

Films of insulating ferroelectric ceramic material have been produced by a variety of methods and on a number of different substrates. However, there have been no investigations of photovoltaic properties. Oikawa and Toda[4] fabricated thick (20-μm) PZT films on fused quartz and stainless steel substrates by electron beam evaporation. The deposition rates were high (0.5 μm/min). Low-frequency Sawyer-Tower loops showed a saturation remanent polarization of 4.2 $\mu C/cm^2$. This value should be compared with the remanent polarization of the bulk ceramic starting material, which was 30 $\mu C/cm^2$. The film material showed a strong dielectric anomaly at the Curie temperature. The coercive field from the Sawyer-Tower measurements was 5.5 kV/cm^2. The ferroelectric properties developed only after annealing at 650°C for several hours.

Schufer et al[5] prepared thin films of barium titanate by radio frequency (rf) sputtering on ceramic superstrates partially coated with platinum. Films 4 μm thick with grain size of 0.2 to 0.4 μm were produced. The films were annealed at 900°C for several hours. Sawyer-Tower circuit measurements of remanent polarization yielded 3 to 6 $\mu C/cm^2$; 6 $\mu C/cm^2$ is a low typical value for the bulk ceramic material. The coercive field (E_c) was about 20 kV/cm^2.

Ishida et al[6] succeeded in rf sputtering ferroelectric PLZT films onto tin oxide coated quartz substrates. The remanent polarization measured by the Sawyer-Tower hysteresis loop method was 4.2 $\mu C/cm^2$ with E_c about 20 kV/cm. These quantities are not well defined because of the softness of the observed loop. The films were 1 μm thick with a grain size of 0.2 μm.

Films of less than 0.1 μm were sputtered into silicon substrate by Park and Granneman.[7] The relatively low dielectric constants of these films (approximately 30) suggest that they may not be ferroelectric. The barium titanate-silicon system has been further characterized by Panitz and Hu.[8]

Polycrystalline ferroelectric PZT-5A films with remanent polarization of about 4 $\mu C/cm^2$ were produced by Constelleno and Feinstein, who used a sputtering technique in which a collimated ion beam, rather than glow discharge ions, effected the deposition of PZT-5A target material

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[4] *Masaru Oikawa and Kohji Toda, Appl. Phys. Lett.,* _29_ *(1976), 491.*

[5] *H. Schufer, H. Schmitt, K. H. Ehses, and G. Kleer, Ferroelectrics,* _22_ *(1978), 779.*

[6] *Malcoto Ishida, Hiroyuki Matsunami, and Tetsuro Tamaka, Appl. Phys. Lett.,* _31_ *(1977), 433.*

[7] *J. K. Park and W. Grannemann, Ferroelectrics,* _10_ *(1976), 217.*

[8] *Junda K. G. Panitz and Cheng-Cheng Hu, Ferroelectrics,* _27_ *(1980), 161.*

onto a substrate.[9] The deposited material was annealed at about 800°C for several hours to produce active films with macroscopic (0.5-μm) grains.

Ferroelectric barium titanate thick films have been produced by annealing layers of deposited powder. Deposition has been by screen printing and electrophoresis.[10-13] As with other methods of deposition, annealing is required[14] to produce polycrystalline (micrometer or sub-micrometer grain size) layers from the deposited materials.

These efforts represent the various techniques that have been used to obtain thin and thick polycrystalline films of barium titanate, PZT, and PLZT materials. Other ferroelectric materials have been prepared in film form. One example is bismuth titanate prepared in semicrystalline form on a silicon substrate by Wu.[14]

5. COMPARISON WITH EAPROM DEVICE

The potential (speed and permanence) of photovoltaic ferroelectric-semiconductor devices can be seen by comparing the predicted characteristics of a photovoltaic ferroelectric programmable device (fig. 3) with a commercially available word alterable EAPROM device of the charge storage type. One such typical device is organized in 1024 four-bit words. A special erase operation erases one or all words simultaneously in 10 ms. Writing then requires 1 ms/word. Block programming a unit therefore requires a time interval in excess of 1 s. Subsequently erasing and reprogramming a single word requires 11 ms.

The photovoltaic ferroelectric-semiconductor EAPROM device (fig. 3) would require a block erase time of about 10 μs. Block programming could be accomplished in as little as 100 ns/cell. In a unit organized similarly to the ER3400, the block programming would require about 100 μs rather than 1 s. Erasing and reprogramming a single word would take 10 μs rather than 10 ms.

Retentivity (unpowered) of the ER3400 is limited to 10 yr at room temperature. Typically, slight elevation in storage temperature reduces

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[9] *R. N. Castelleno and L. G. Feinstein, J. Appl. Phys.,* *__50__ (1979), 4406.*

[10] *Vernon Lamb and Harry I. Salmon, Ceramic Bull.,* *__41__ (1962), 781.*

[11] *J. Dec and Z. Surowiak, Phys. Papers, Silesian University, Katowice,* *__6__ (1978), 74.*

[12] *Z. Surowiak, Acta Phys. Pol.,* *__43__ (1980), 543.*

[13] *M. Loposzko and Z. Surowiak, Thin Solid Films,* *__67__ (1980), 253.*

[14] *S. Y. Wu, IEEE Trans. Electron. Dev.,* *__ED-21__ (1974), 499.*

markedly the unpowered storage time. The ferroelectric device is expected to store information for an indefinitely long time even at elevated temperatures.

The information content of the commercial charge storage ER3400 can be expected to be removed by a burst of penetrating ionizing radiation. This is not expected for the ferroelectric devices in which the information is stored in remanent polarization. This storage mechanism is insensitive to radiation.[1]

The data sheet of the ER3400 gives a maximum of 100,000 for erase/write cycles. The test data for the ferroelectric test cell, on the other hand, show proper operation after 1.8×10^9 write/rewrite cycles.

Most importantly, no interaction appears between read cycles and stored information in the ferroelectric device. Once programmed, a cell can be accessed at any rate consistent with the 200-ns read time used for the test device and for any length of time without removing the programmed information. There is no read limitation. The ER3400 has a read limitation of about 2×10^{11} read cycles.

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[1] *D. C. Burtoot and G. W. Taylor, Polar Dielectrics and Their Applications, University of California Press, Los Angeles (1979), 299.*

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