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Experiment F03
The Remanent Hysteresis Loop

Objective:
Previously, we learned how to execute the half-hysteresis loop. In this experiment, we will use this new tool to measure the "remanent hysteresis loop", the shape of the memory in the ferroelectric capacitor.

Method:
We will do half-loops with and without the remanent polarization switching during the loop. We will then subtract a non-switching half-loop from a switching half-loop to derive the switching dynamics of the remanent polarization.

Theory:

System Analysis
We will derive the shape of the hysteresis of the remanent polarization in our ferroelectric capacitors using a technique called "systems analysis". Basically, systems analysis involves stimulating an *unknown* system with a *known* input and measuring its output to find the *transfer function* of the system. The transfer function for a system is defined as the mathematical function applied by the unknown system to the input in order to generate the output.

$$\text{Transfer Function(stimulus)} = \text{system output} \qquad \text{eq(F03.1)}$$

Dividing eq(F03.1) by the stimulus gives the transfer function for that system:

$$\phi = \frac{\textbf{System output}}{\textbf{Stimulus}} \qquad \text{eq(F03.2)}$$

Those of you who have degrees in electrical engineering will remember systems analysis as mostly concerning the performance of Fourier transforms on the output of a system to which you applied a Dirac delta function as an input. For those of you who are not engineers or scientists, Fourier mathematics holds that any continuous signal, even a square wave, can be constructed from the sum of selected sine waves. The Fourier transform is the reverse operation in mathematics that tells you which sine waves and their amplitudes from which a signal is constructed. Finally, a Dirac delta function is a theoretical input signal containing sine waves of all possible frequencies from zero to infinity. In a nutshell, systems analysis stimulates a system with all possible sine waves, measures which ones come out the other end, and then derives the transfer function from the results! (Theoretically, you can now skip a whole semester of electrical engineering classes!)

> Advanced Concepts: Jean Baptiste Joseph Fourier (1768-1830) created one of the most powerful mathematical tools every conceived by man. Its importance to quantum mechanics, information theory, control theory, systems analysis, optics, instrumentation, aeronautics, physics, astronomy, chemistry, and measurement theory cannot be overstated. Our modern society would not exist without the tool he created. There are many textbooks teaching Fourier analysis. A nice text in which to understand Fourier and his mathematics in their historical context is God Created the Integers, edited by Stephen Hawking [Running Press, Philadelphia, 2005, pp491-499]. Hawking examines a wide range of notable mathematicians in addition to Fourier, going all the way back to Euclid and Archimedes. Ninety percent of the book is dense reading as Hawking reproduces original writings from each mathematician. Ancient Greek mathematics translated into English cannot be skimmed easily. Nevertheless, the remaining ten percent of the book is intriguing as Hawking details the personal histories of the mathematicians, placing their lives and creations in context of the times in which they lived.

We have already unknowingly executed systems analysis in our study of the linear capacitor in Experiment L01.

$$Q = C \bullet \text{Volts} \qquad \text{eq(1.3)}$$

$$C = \frac{Q}{V} = \frac{\textbf{System Output}}{\textbf{System Input}} \qquad \text{eq(F03.3)}$$

C, capacitance, is the transfer function between the voltage across the capacitor and the charge the capacitor generates.

> Note: for those if you who are who are classically trained, which is just about everybody, the transfer function for a capacitor is

$$Z = \frac{1}{2\pi\omega C}, \qquad \text{eq(F03.4)}$$

an equation which is true over all frequencies. A unique property of the EDU is that it operates in an *impedance-free* mode, making ω irrelevant and allowing us to revert to the much simpler eq(F03.3) without error as long as we stay within the specified frequency envelope of the EDU.

A second important construct in systems analysis is the assumption that a complex system can be constructed from a series of independent linear systems. The output of the complex system will be the *sum* of the responses of all the simple systems making up the complex system. Each simple system in the complex system is called a *component* of that complex system and the assumption means that each component operates on the stimulus *independently* of the other simple systems making up the complex system.

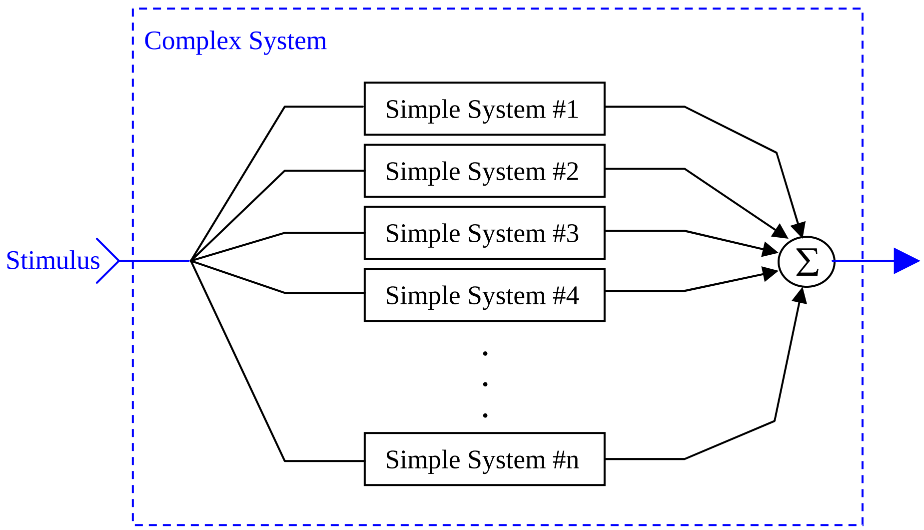

Figure F03.1
Components of a Complex System

Components of a system such as that shown in Figure F03.1 are said to have the property called *superposition*. None of the components affect any of the others so the output of the system is the simple sum of the outputs of the components.

Linear systems such as linear capacitors make classical tools behave well in systems analysis. Applying a single sine wave to linear *simple system* causes the output sine wave to be related to the input sine wave by a constant. The response of the same system to a Dirac delta input will be an array of constants, one for each sine wave frequency in the stimulus/output signal. As we have discussed throughout the tutorials and the experiments for the EDU, the output of a linear system *looks just like the input but with a different scale.* The ratio of the scales is the transfer function. The Polarization vs Time plot of the hysteresis loop of a linear capacitor in the figure below demonstrates this concept.

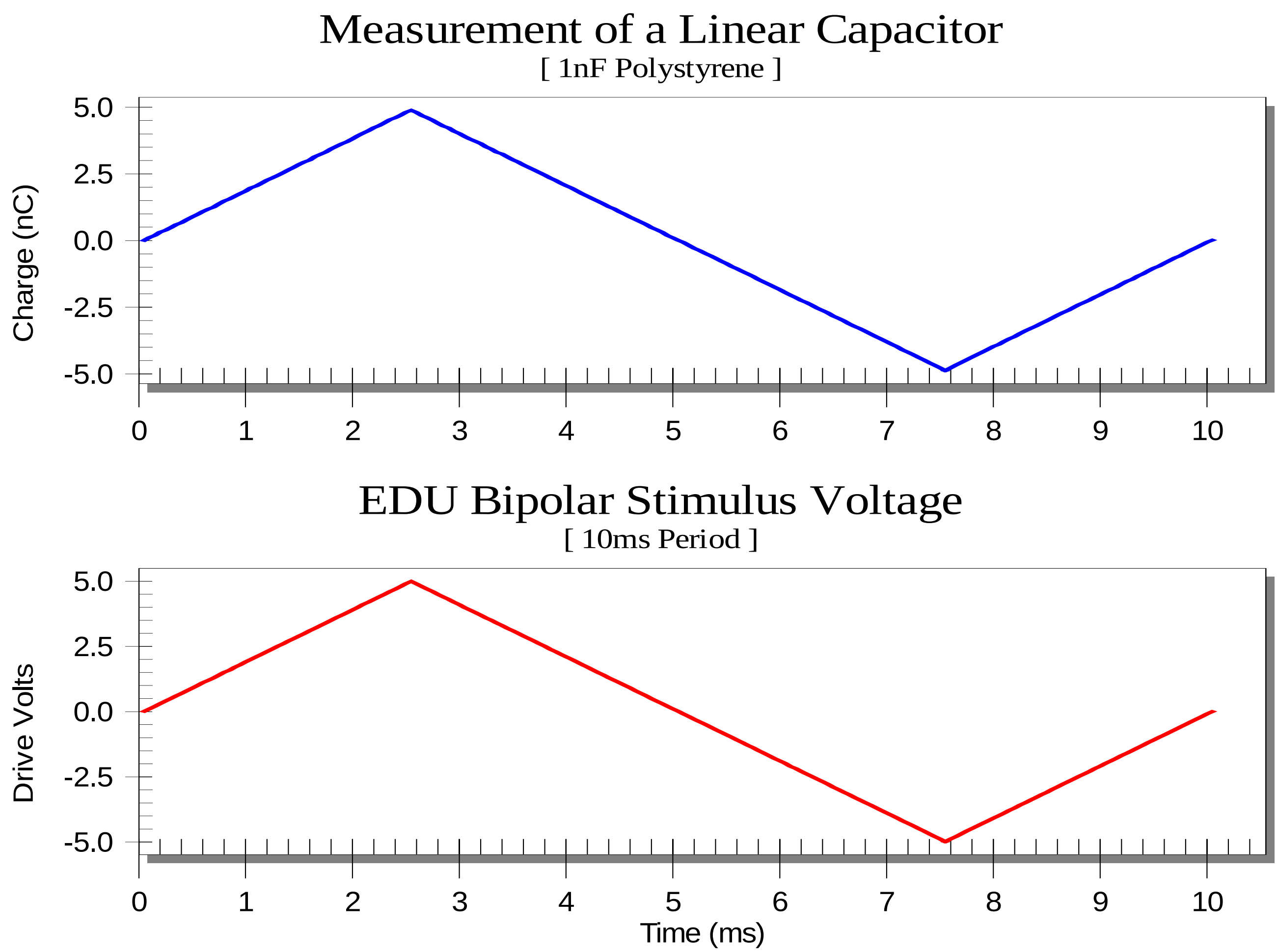

Figure F03.2
In a linear system, the output looks like the input.

In a *non-linear simple system* stimulated with a single sine wave of a single frequency, some of the amplitude of the input sine wave is transferred to a sine wave of a different frequency! Where one sine wave is input into the simple system, more than one sine wave comes out of the system. In other words, the output does not look like the input. The more non-linear the system is, the greater the deviation of the *shape* of the output from the input. The hysteresis of a ferroelectric capacitor in Figure F03.3 below quite clearly indicates its non-linear nature.

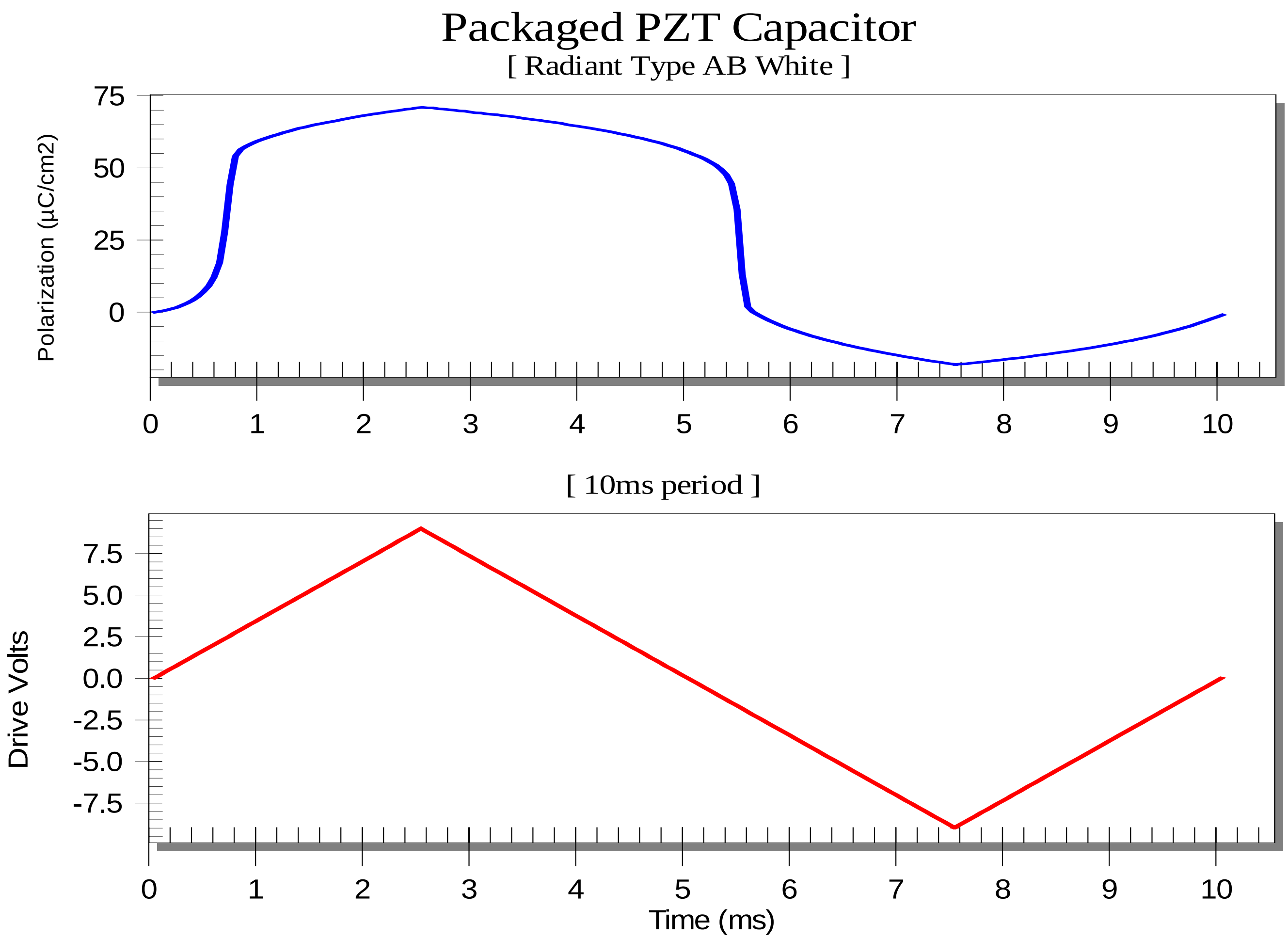

Figure F03.3
In a non-linear system, the output may look nothing like the input.

You can reproduce Figure F03.3 from your data by *recalling* any of the hysteresis loops of a ferroelectric capacitor that you have in your data folders and using the PLOT button to re-graph the data as "Polarization vs Time".

A non-linear system may also not meet the principle of *superposition*. In such a system, the output of one component of the complex system affects the inputs or outputs of another component.

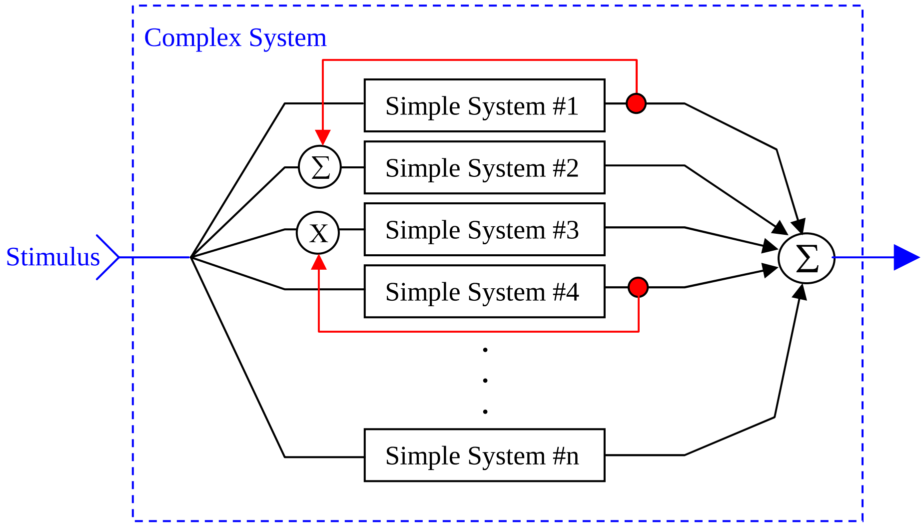

Figure F03.4
A Non-Linear Complex System with Feedback

The **red** paths in Figure F03.4 are called *feedback* loops. They will modify the output of the entire system not only based on the input stimulus but also on the functional relationships between the different simple systems. Feedback loops can make the entire system go unstable under the proper stimulus conditions but they are very important in our technological society. Feedback is the subject of study in the discipline of *control theory*.

In the non-linear complex system of Figure F03.4, the functions themselves do not change. Another type of relationship between the components of a complex system occurs when the output of one component *modifies the transfer function* of another component.

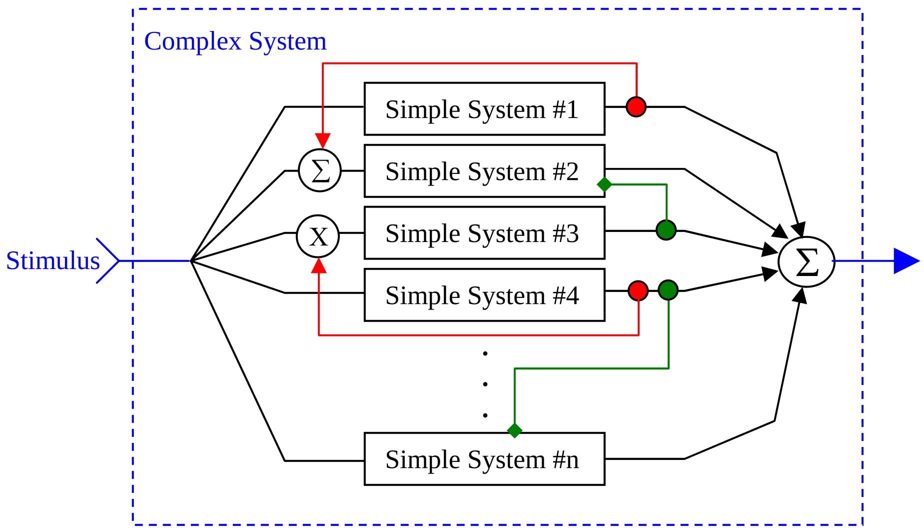

Figure F03.5
A Non-Linear Complex System with Feedback and Transfer Function Modification

The **green** lines in Figure F03.5 indicate paths by which the outputs of some components feed into and modify the transfer function of another. This type of complex system is very common in nature. For an intimidating example of component transfer function modification in a complex system, pick up any biochemistry textbook and attempt to map out the control paths for an intracellular function. A good example is the affect of insulin on the molecular functions inside a cell that regulate the uptake of sugar by the cell as well as the bewildering array of molecules outside the cell that affect the function of insulin.

All three of the system architectures described above operate in ferroelectric materials during a hysteresis measurement. The electric fields of a unit cell dipole will move so as to cancel any electric field applied externally to the cell, reducing the strength of that external electric field. This is a feedback mechanism operating at the microscopic level. The electric fields of dipoles in a ferroelectric material apply a force against all of the other dipoles in the material, modifying the transfer function of each dipole and hence the transfer function for the entire capacitor. (This residual force also changes the physical size of the capacitor!) Finally, the hysteresis loop is the superposition of the state of all of the unit cells in the material lattice after they have been modified by feedback and force, an implementation of the superposition principle. With trillions of unit cells in even a small capacitor, these interactions get complicated very fast. As well, the stimulus signal for measuring the ferroelectric hysteresis loop is not continuous. It starts and stops in time. This discontinuity greatly complicates the representation of the stimulus signal in Fourier mathematics. For these reasons, *there exist today no formal mathematical* *descriptions of the function of ferroelectric materials that are accurate over the range of the microscopic to the macroscopic levels and over short to long time scales.*

System Analysis through Isolation

How, then, can we analyze the ferroelectric hysteresis loop for internal components? Does it even have components acting in superposition with each other?

First, we will not attempt to convert the measurements from absolute values to the sum of mathematical functions. The envelope of the measurement, its shape, will carry the information we seek. We will thus treat each measurement as a single set of information to be added or subtracted point-per-point from other sets much like we subtract "B" from "A" in algebra. Instead of mathematically identifying the different components, we will *isolate* a particular component in a measurement so that it does not contribute to the result of the measurement or so that it becomes the only component that contributes to the output signal of that stimulus. We can then add or subtract the measurements to pull out the components that cannot be measured directly. Just like the rule in algebra about needing as many equations as there are variables to find the value of all variables, *to complete our systems analysis we will need as many independent measurements as there are components in the hysteresis loop*.

For this experiment, we will attempt to measure the remanent polarization of the ferroelectric capacitor. We will assume a model of the hysteresis consisting of two components: remanent polarization and all other polarizations that one would expect to see in a non-ferroelectric capacitor. In reality, there are at least four components and probably more but we will stick with only two for this experiment. Consequently, we will need to make two independent measurements of the capacitor in a way that isolates the remanent polarization.

The remanent polarization is that charge which remains behind once a voltage stimulus returns to zero. We know from our theoretical knowledge of the material that this remanent polarization will switch in a direction to cancel an applied voltage and that the remanent charge held on the plates will move during the measurement. The theory also means that the remanent polarization *will not switch* if the voltage is applied in the same direction again. This definition is easy to visualize. Any polarization that does "switch back" while the stimulus returns to zero is not remanent. Therefore, we have our two independent measurements: 1) one with remanent polarization switching and 2) one with no remanent polarization switching. We can subtract the two to see the remanent polarization. Our component model is then

Switching Half-Loop = Remanent Polarization + Non-switching Half-Loop eq(F03.5)

If we assume superposition of the components, then

Remanent Polarization = Switching Half-Loop - Non-switching Half-Loop eq(F03.6)

We are guaranteed to get the full remanent polarization on the switching half-loop if we do a saturated half-loop in the other direction first. Consequently, the test will look as follows:

Step #1: Switching half-loop ***against*** the test direction (-9V)

Step #2: Switching half-loop ***in*** the test direction (+9V)

Step #3 Non-switching half-loop ***in*** the test direction (+9V)

The component can then be found using the following equation:

Remanent Polarization = Step #2 - Step #3 eq(F03.7)

Step #1 does not contain useable information. It is necessary, though, to ensure that Step #2 starts with all of the polarization opposite the direction of test. The applied signal in Step #1 is called the *preset loop* and all research level ferroelectric materials testers execute one prior to measuring the hysteresis loop for presentation.

Verifying the Accuracy of Superposition

The test procedure outlined above *assumes* that the remanent polarization component exhibits *superposition* with all other components present in the measurement of the ferroelectric capacitor. Otherwise, the measurements in both Step #2 and Step #3 would contain remanent polarization and the difference calculated in eq(F03.7) would not be the complete remanent polarization. How can we verify that this assumption is reasonable if we cannot apply classical mathematical analysis to the results?

Fortunately, we can predict attributes that the remanent polarization should have. If our results have these attributes, then we can reasonably assume that our component model using superposition is correct despite that fact that internal to the capacitor, no unit cell is independent of another.

First, memory has a unique characteristic in that it does not change after being set as the stimulus *returns to zero*. Otherwise, it would not be part of the memory component. The rule may be expressed as

$$\Delta\text{Memory} = 0 \text{ as the stimulus returns to zero} \qquad \text{eq(F03.8)}$$

or, for our ferroelectric capacitor,

$$\Delta\text{Memory} = \frac{\Delta Q}{\Delta V\downarrow} = \text{Capacitance} = 0 \qquad \text{eq(F03.9)}$$

In other words, the trace of the remanent polarization as the voltage goes from Vmax to zero should be flat! Another way to state it is that the capacitance of the retrace is zero. This is a quality factor we can easily evaluate on our results.

Second, there are two states we are allowing in our experiment for memory in the ferroelectric capacitor: 1) all of the dipoles facing UP and 2) all of the dipoles facing DOWN. Assuming that all of the dipoles participate equally in both the UP and DOWN states, the remanent polarization measured in the positive direction and in the negative direction should be equal.

$$+\text{Premanent} = -\text{Premanent} \qquad \text{eq(F03.10)}$$

The remanent polarization in the negative direction can be measured by inverting the voltages listed for the positive measurement. The experiment then becomes

Step #1: Switching half-loop ***against*** the ***positive*** test direction (-9V)

Step #2: Switching half-loop ***in*** the test ***positive*** direction (+9V)

Step #3 Non-switching half-loop ***in*** the ***positive*** test direction (+9V)

Step #4 Switching half-loop ***in*** the ***negative*** test direction (-9V)

Step #5 Non-switching half-loop ***in*** the ***negative*** test direction (+9V)

Notice that we could have generated a test procedure with six steps by simply repeating the three test steps in the opposite direction. The five-step test above eliminates one step by using the non-switching positive test (Step #3) as the preset for the switching negative test (Step #4). This five-step test procedure is called the *PUND test* in the ferroelectric community.

In summary, we will attempt to measure the remanent polarization hysteresis of a ferroelectric capacitor assuming the superposition principle of systems analysis. We have two quality factors with which to confirm the accuracy of our assumption of superposition:

1) The retrace of the remanent hysteresis loop should have no capacitance

and

2) the positive and negative going measurements should return the same total remanent polarization value.

> Advanced Concepts: For those who research ferroelectric materials for a living, the model is much more complex than presented here. While the superposition principle applies well enough to give good results for our experiment below, the components of the ferroelectric capacitor are not independent of each other. Remanent polarization modifies both the small signal capacitance and the resistive leakage of the device. Consequently, the shape of the switching half-loop is not exactly the same as the shape of half-of-a-continuous hysteresis curve. As well, there are parasitic components that decay after stimulus so their contribution to the hysteresis loop changes as the test frequency changes. If the test is slow enough, the parasitic components decay totally during the test and the loop looks more square than if the test is fast and the parasitics do not have time to decay before the loop is over. The controversial "gap" in the hysteresis loop is one consequence of these parasitic effects. Another issue arises from permanent changes in the remanent polarization between tests or during tests. Fatigue operates per "log(cycle)" so PUND tests executed on virgin capacitors may show some difference in the positive and negative going remanent polarization. The same PUND test executed after the capacitor has seen 1000 cycles will show no difference between the two remanent polarization measurements. Nevertheless, the total polarization measured in the later tests will be lower due to the fatigue arising from the earlier cycles. Finally, imprint of the hysteresis loop with time-in-state will cause asymmetries in the remanent polarization measurements.

For those of you just being introduced to ferroelectrics, do not worry about the non-linearities I describe in the preceding "Advanced Concepts" paragraph. They will not affect our test to any significant degree. You can study retention, fatigue, and imprint in later experiments if you desire. Researchers in ferroelectric materials *must worry* about these properties in their tests because their job is to design commercial products where these materials must withstand billions of fatigue cycles and extensive temperature cycling over one or more decades without losing their functionality. Under those circumstances, the little things count.

Procedure:
We will recover our test capacitor to make sure it is symmetrical. We will then select the "monopolar" test option and make the five half-loop measurements required for our experiment.

1) Execute a recovery procedure on a type AB White capacitor.

2) Open the CONFIGURE TEST menu and set the following conditions:

   a) Vmax = -9V

   b) Hysteresis Period = 10ms

   c) Driver Profile Type = Standard Monopolar

   d) Type in the name of your sample in the "Sample Name" window.

   Go to the PLOT SETUP page of the menu.

   e) Data Label = "Preset Half-Loop".

   f) Plot Title = "Switching vs Non-switching Half-Loops"

   g) Plot Subtitle = [Radiant Type AB Cap]

3) Close the menu and execute the test. CLEAR DATA. (We do not need this data.)

4) Open the CONFIGURE TEST menu again.

   a) Change Vmax to +9V

   b) On the PLOT SETUP PAGE, change the Data Label to
      "Switching Half-Loop UP"

5) Close the menu and execute the test.

6) Press the STORE DATA button and save the data.

Name the file "Switching Half-Loop UP". Save the file in a new folder entitled "Experiment F03 Folder".

7) Open the CONFIGURE TEST menu again.

a) On the PLOT SETUP PAGE, change the Data Label to "Non-switching Half-Loop UP"

8) Close the menu and execute the test.

9) Press the STORE DATA button and save the data.
Name the file "Non-switching Half-Loop UP". Save the file in the folder "Experiment F03 Folder".

Your data plot should now contain the switching and non-switching half-loops in the positive direction.

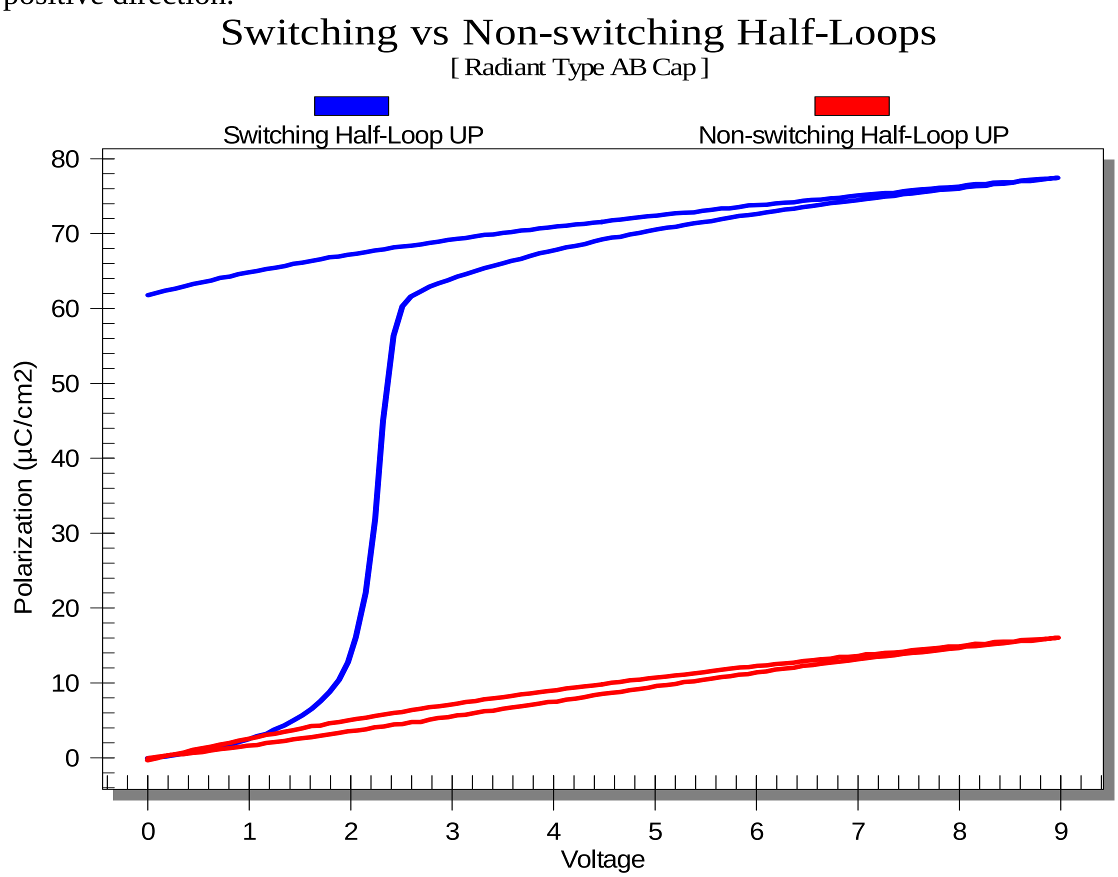

Figure F03.6
The Positive Switching and Non-switching Half-Loops

10) Open the CONFIGURE TEST menu again. Change Vmax to –9V and the Data Label to “Switching Half-Loop DOWN” .

11) Execute the test and save the results in the file “Switching Half-Loop DOWN” in the “Experiment F03 Folder”.

12) Finally, open the CONFIGURE TEST menu one last time. Leave Vmax equal to –9V but change the Data Label to “Non-switching Half-Loop DOWN” .

13) Execute the test and save the results in the file “Non-switching Half-Loop DOWN” in the “Experiment F03 Folder”.

If you have not erased your plot between each test, you should now have all four measurements in a single graph. The difference between the switching and non-switching loops is readily apparent, one reason we chose 20/80 PZT for the EDU. Also note the importance of the data labels for identifying the loops.

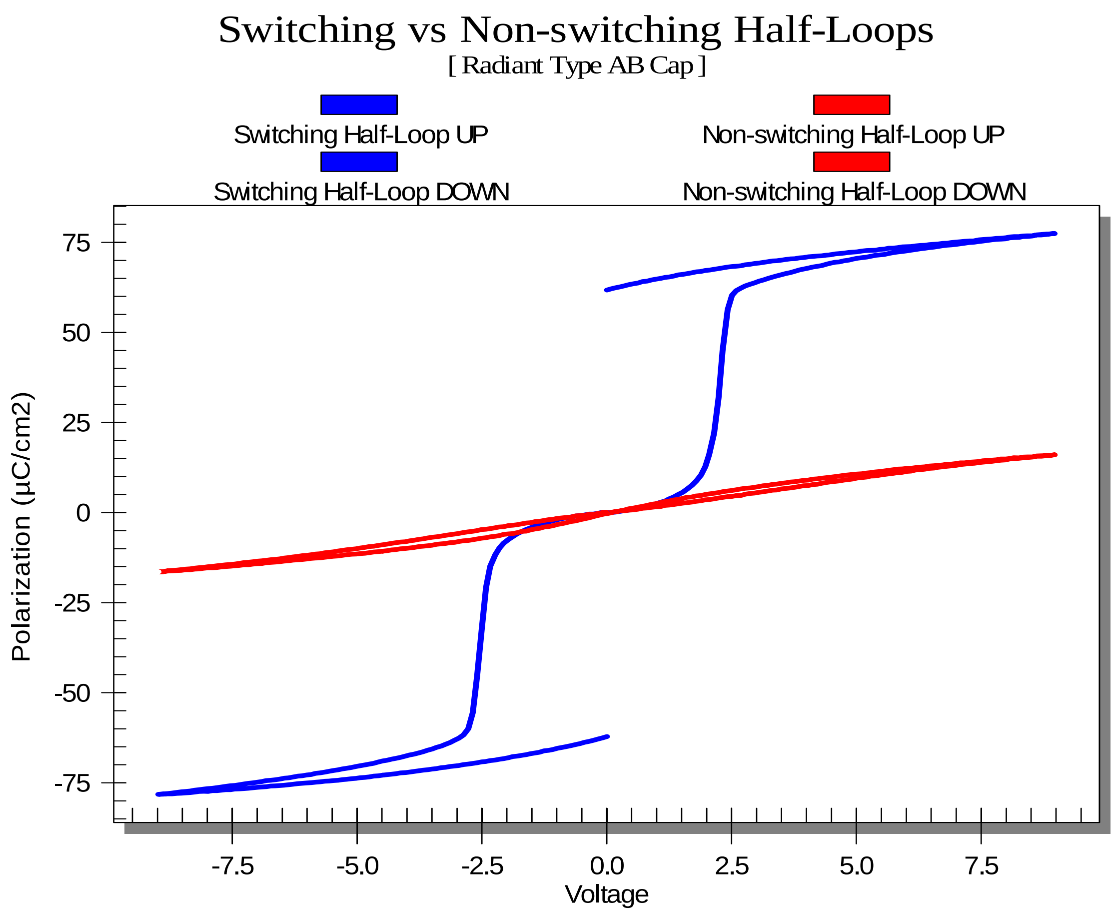

Figure F03.7
All Four Switching and Non-switching Half-Loops

All of the data is now collected. We can proceed with the derivation of the remanent polarization hysteresis.

14) Erase the plot. One at a time, recall each of the files from the "Experiment F03 Folder"

"Switching Half-Loop UP"
"Non-switching Half-Loop UP"
"Switching Half-Loop DOWN"
"Non-switching Half-Loop DOWN"

and, using the pop-up menu, export them direclty from the graph in ASCII format to a spreadsheet using the technique described in Experiment I04 – Plotting Tools.

IMPORTANT: Save the spreadsheet file in the "Experiment F03 Folder" so it is always physically present with the EDU data files.

15) Using the spreadsheet tools, calculate the following functions and plot them together on a single graph.

+Premanent = (Switching Half-Loop UP) – (Non-switching Half-Loop UP)

-Premanent = (Switching Half-Loop DOWN) – (Non-switching Half-Loop DOWN)

My remanent polarization half-loops are plotted below:

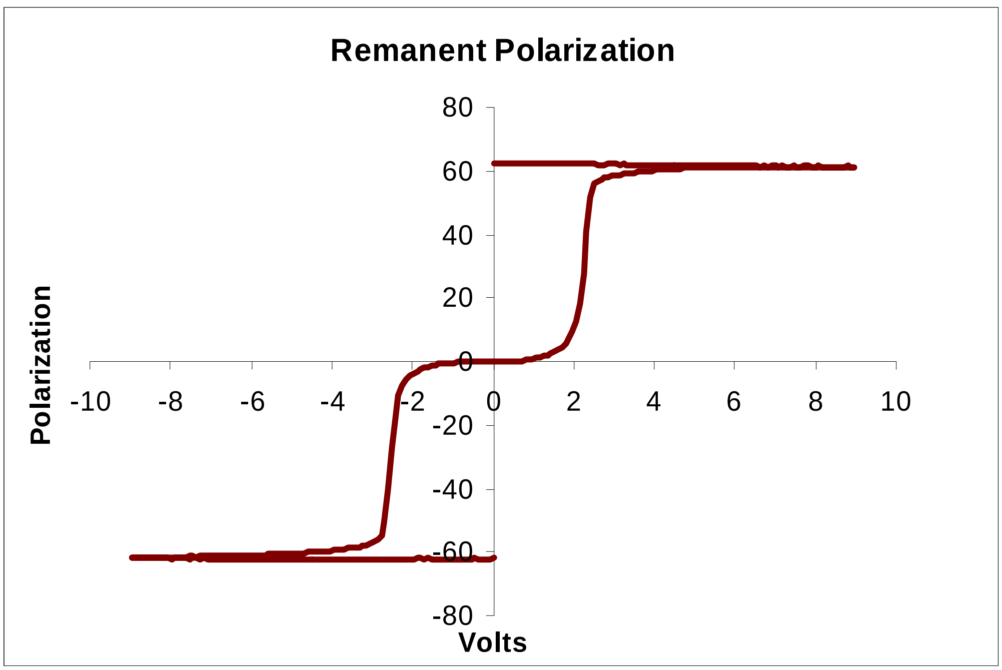

Figure F03.8
The Positive and Negative Remanent Polarization Half-Loops

There is one more operation we must perform. We must manually center the two remanent half-loops so we can compare their amplitudes. To do this, find the average of the first and last point of the positive remanent half loop and subtract that average from every point in the positive loop. Do the same for the negative loop. This operation should move the positive loop down in the plot so its first and last points are equally split vertically around the X-axis. The negative remanent loop should move up the same way. On a well-behaved sample, the positive and negative remanent half-loops will appear to merge after centering as if they form a single measurement taken in a single pass. As well, the retrace segments of both half-loops should be flat.

> NOTE: If the retrace segments are not flat, repeat the experiment. If they are not on the second try, then your capacitor is changing *as you take the data.* This effect is discussed in the Advanced Concepts paragraph above.

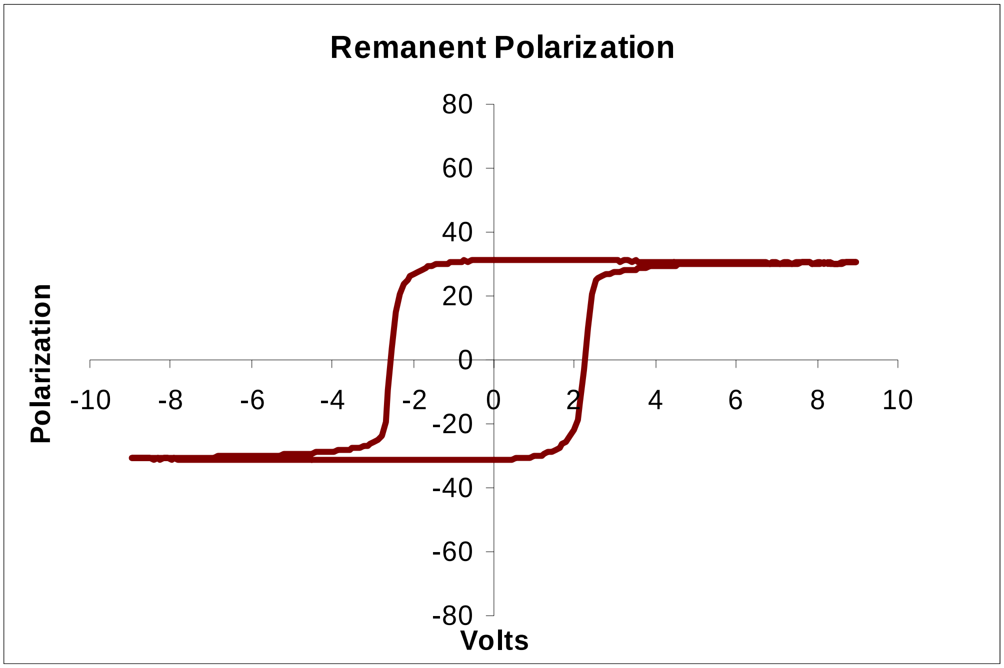

Figure F03.9
The Remanent Polarization Loop

Now that we have the centered the positive and negative remanent half-loops, we can apply our two quality factors to determine if our model of the system is valid.

Quality Factor #1: Are the retrace segments of both loops flat? *Yes!*

Quality Factor #2: Is the remanent polarization the same measured in the positive and negative direction? *Yes! The two loops appear to merge into a single loop.*

Conclusion to Experiment F03

Experiment F03 was fun. We successfully derived the hysteresis loop of the remanent polarization using the PUND format combined with half-hysteresis loops. It appears that our model of the ferroelectric capacitor is correct. We can separate the hysteresis into two nominally independent components: 1) the remanent polarization and 2) the other capacitor polarization components. It is important that you consider the meaning of the remanent polarization loop. It represents the internal *memory* state of the capacitor and it has a profound applications in memory devices, sensors, and actuators. In the next experiment, we will further study the remanent polarization. Does it have only two states, UP and DOWN, or can it take on states in between?

Recommended Exercise:

What is the relationship between the remanent polarization and the full hysteresis loop? To study this, continue your measurements of this same capacitor to capture a full positive going hysteresis loop (Vmax = +9V) and full negative going hysteresis loop (Vmax = -9V) using the Standard Bipolar format. Export the two new loops and plot them together with the centered remanent polarization loops we captured in this experiment. How are they related? Are the full loops continuous or do they have gaps in t hem? What is the relationship between the gaps in the full loops and the remanent polarization loop? Can you generate a general rule for deriving the remanent polarization value using on full loops?

> NOTE: Perform this exercise in one sitting. If you wait too long between the different parts of the measurement, the imprint mechanism will change the results. The 20/80 PZT used in the Type AB capacitors makes beautiful square hysteresis loops. However, it has strong fatigue and imprint characteristics, strong enough to see in this experimental format.