Resonant Piezoresponse Force Microscopy

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CHAPTER 1
INTRODUCTION

1.1 Background

An increasingly popular method for local ferroelectric characterization is by piezoresponse force microscopy (PFM)[1], a variant of atomic force microscopy. In this technique, a conductive tip is placed in contact with an electromechanically activate material and contact is maintained through a control feedback. A laser is focused onto the back of the tip and the reflection of the laser is collected with a photodetector. Then, an sinusoidal electric field is applied to the material through the tip and the material strains due to the converse piezoelectric effect. Ideally, the frequency of the applied field is the resonant frequency of the tip-surface interactions, increasing the response and decreasing noise. The deflections of the tip and, thus, the material strain is collected via the photodetector. The amplitude (A_0) of the deflections correspond to the piezoelectric coefficient and the polarization direction of the material (ϕ) can be obtained from the phase difference in the applied voltage and the measured deflections.

In this way PFM maps the local electromechanical response with a spatial resolution that roughly corresponds to tip radius (at best on the order of $10\ nm$). This approach has been widely used to study mesoscale domain configuration of FE materials and has informed on mesoscale phase transitions[2], as well as domain configuration dependence on chemistry and temperature[3, 4, 5, 6, 7]. Additionally, PFM is capable of probing the electromechanical response as a function of time and/or voltage, enabling measurement of local switching behavior or other time/field-dependent electromechanical behaviors[8, 9]. Further, by applying these measurements over a surface area in a grid the evolution of various behaviors, such as ferroelectric hysteresis, have been spatially mapped on the mesoscale with nanometer resolution[10, 9, 11].

However, traditional PFM is limited by the fact that it is ”single-frequency”, i.e. the material response is only probed at one frequency. Thus, significant tip-surface resonance shifts, which can be caused by surface topography and phase transitions[12], can result in material response being completely lost to noise. Further, probing only a single frequency results in an under-specified system. For instance, a common assumption is that the tip-surface interactions are well-described by a simple harmonic oscillator (SHO):

$$f(\omega) = \frac{A_0 e^{i\phi} \omega_0^2}{\omega^2 - \frac{i\omega\omega_0}{Q} - \omega_0^2} \tag{1.1}$$

Where A_0 is the amplitude at resonance, Q is the quality factor, ω_0 is the resonant frequency, and ϕ is the phase. With this *a priori* assumption, it becomes clear that single frequency PFM, yielding only the amplitude and phase, does not specify a unique solution for the system and this ambiguity leads to spurious image contrast[13, 14].

Numerous extensions to PFM have been developed to measure the frequency dependence of the response to alleviate these issues. Techniques like dual AC resonance[15] employ a control scheme to follow resonance, while others seek to capture the response–frequency behavior over a wide band of frequencies. These approaches include fast lock-in sweeps[16], pulse excitation[17, 18], and band excitation[19]. Among them, band excitation (BE) stands as a particularly attractive option due to a comparatively faster measurement rate, higher signal intensities, and reduced topological cross-talk[20]. In BE-PFM, the response–frequency behavior is probed by exciting the material with a band of frequencies, measuring the response, and using a functional fitting to Eq. 1.1. However, currently available BE-PFM systems are either proprietary and only available at user facilities or only available as purchasable upgrades specific to particular PFMs. Further, in some implementations spectroscopic BE-PFM measurements suffer from a strict limit on acquisition time, thus impeding measurements that require higher than average data points. Here, a fully open-source wide bandwidth system was developed independent of any AFM, rendering the system applicable to a wide range of AFMs. Further, a supplementary open-source tool for handling, processing, and plotting the resulting data was developed.

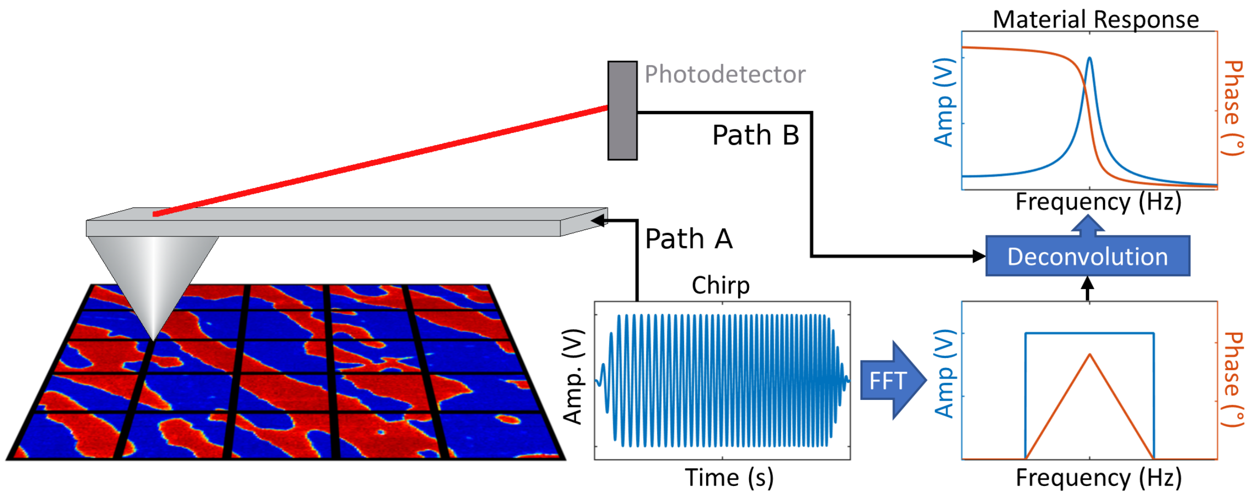

Figure 2.1: Illustration of resonant piezoresponse force microscopy (R-PFM) for application of a single chirp. First the excitation waveform is generated according to user specifications and applied to the material through the tip. Then, the AC deflections of the tip are collected from the photodetector. Finally, the AC deflections are deconvolved with the applied chirp and the material's response is obtained.

CHAPTER 2
THEORY AND REQUIREMENTS

Resonant PFM (RPFM) is the generation and application of a band of frequencies and subsequent collection of the material response within a reasonable time frame. Thus, RPFM can be conceptually divided into two "paths": the application path (Fig. 2.1 path a), which applies a waveform, and the collection path (Fig. 2.1 path b), which collects the response of the material to the applied waveform and extracts parameters from a fitting to Eq. 1.1.

2.1 Application path

Strictly speaking, in RPFM (and any wide band probing technique) only the excitation or driving waveform is necessary to probe the material response (Fig. 2.1 path a). However, important information on the ferroelectric properties of materials is obtained from DC-field and/or time dependent probings, such as in switching spectroscopy and relaxation measurements. Thus, a necessary functionality of RPFM is to overlay the excitation waveform with an arbitrary and user-definable waveform (e.g. Fig. 4.2), enabling such electric field based spectroscopy. Here, we will first discuss the excitation waveform and then move onto how the excitation waveform can be overlaid an arbitrary waveform to enable spectroscopic probings.

2.1.1 The Excitation Waveform

In single frequency PFM, the material is excited with a single frequency sinudsoid. The fundamental idea behind RPFM is to instead excite the materials with a *band* of frequencies. Thus, the ideal frequency spectrum of a RPFM excitation waveform is simply a constant amplitude within the desired band of frequencies with an amplitude of zero outside the band, illustrated in Fig. 2.1. In the RPFM system described here, the excitation waveform is based on a linear swept-frequency sinusoidal (or ”chirp”)[21], defined as:

$$x(t) = A_0 sin((f_0 + kt)t) = sin(f_0 t + kt^2) \tag{2.1}$$

Where t is time, f_0 is the initial frequency, A_0 is the amplitude, and k is the sweep rate (also known as chirpyness). This chirp waveform indeed exhibits the desired frequency response. However, ringing in the frequency response occurs when the chirp is finite in time, due to the discontinuities occurring at the beginning and end of the waveform. This ringing can be minimized or eliminated by tapering the chirp, which gradually reduces the amplitude at the beginning and ending of the chirp. Thus, the final excitation waveform used here is a chirp tapered via a Tukey window[22], Fig. 2.2 provides an illustration.

The user dictates the excitation chirp by specifying: amplitude A, lower frequency f_L, upper frequency f_H, sampling rate f_s, and chirp duration Δt. The chirp is calculated by determining the parameters for Eq. 2.1 from these user specified parameters as shown in Eq. 2.2 and the chirp is tapered.

$$A_0 = A \tag{2.2a}$$

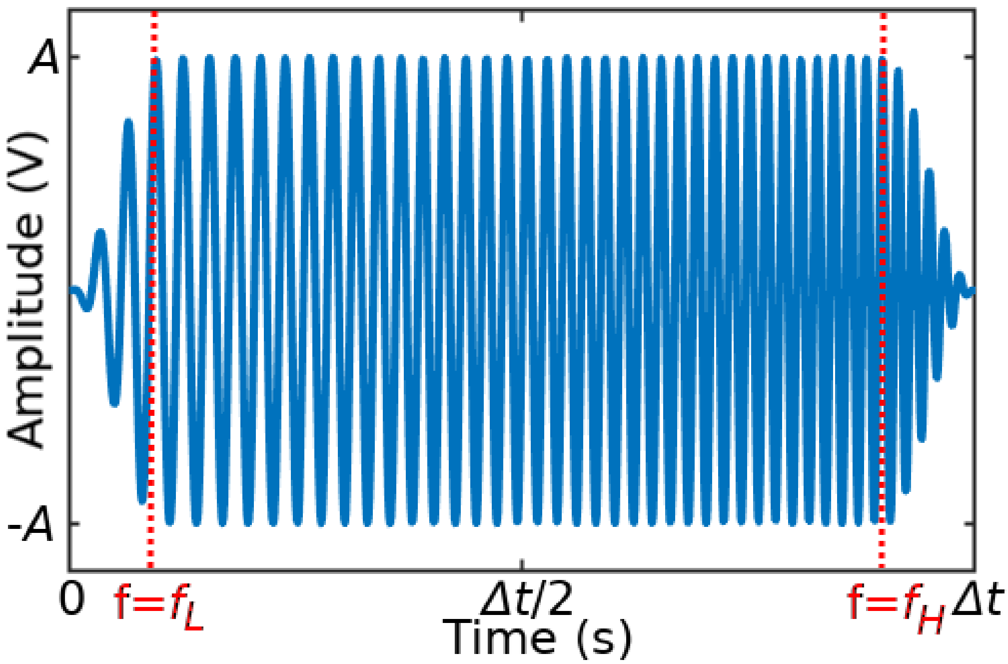

Figure 2.2: Illustration of the excitation waveform, or chirp, annotated with the user specified parameters. The chirp is calculated from the user-specified chirp amplitude (A), lower and upper frequencies (f_L and f_H), sample rate, and chirp duration (Δt). Here, A is the peak value of the chirp, Δt is the time length of the chirp, and f_L and f_H are the desired low and high frequencies of the chirp. The specified f_L and f_H are automatically adjusted to account for the windowing of the chirp, which reduces the amplitude at the beginning and ending of the waveform.

$$f_0 = 0.98 * f_L \tag{2.2b}$$

$$k = \frac{1.02 * f_H - 0.98 * f_L}{\Delta t} \tag{2.2c}$$

Note that in Eq. 2.2, the low frequency is decreased by 2% and the high frequency is increased by 2%. This is to account for the tapering and maintains a constant amplitude throughout the user-specified band. What follows is an in depth description of each parameter, with an illustration of the parameters in Fig. 2.2.

- *Chirp Amplitude:* The Chirp Amplitude is the peak voltage of the chirp (A_0 in eq. 2.1) and should be specified in volts.

- *Lower and Upper Frequency:* The Lower and Upper Frequency are the frequencies bounds of the measurement (f_L and f_H in eq. 2.1), specified in Hz. **Note that the user specified frequencies are automatically adjusted by 2% to account for the windowing, as illustrated in eq. 2.2 and Fig. 2.2**.

- *Sampling Rate:* The Sampling Rate is frequency at which the waveform is generated and the response sampled (f_s in eq. 2.1), specified in Hz, and is dictated by Nyquist-Shannon sampling theory.

- *Chirp Duration:* The Chirp Duration is the time length of the excitation waveform, in seconds. This parameter is the defacto time step of the entire measurement, as a functional fitting is ultimately applied to the frequency response of individual chirps to extract parameters. An important relationship to consider here is the inverse relationship between the time step and frequency resolution of the ultimately obtained frequency response (i.e. material response in Fig. 2.1), or $\Delta f = \frac{1}{\Delta t}$. Δf must be reasonably small to produce an adequate fit.

2.1.2 Electric field and time dependent spectroscopy

While it is the RPFM excitation waveform (or chirp) that probes the material response and is the core of RPFM, a means of field- and time- dependent probings is necessary for a functional PFM system. Typical PFM based spectroscopic probing can be roughly categorized into DC-field dependent measurements, AC-field dependent measurements, and time-dependent measurements. For instance, switching spectroscopy would be considered DC-field dependent measurements, while electromechanical non-linearity measurements would be AC-field dependent. Further, often the higher order harmonics of the electromechanical response is investigated to inform on phenomena such as electrostriction. Thus, to fully enable common spectroscopic measurements a RPFM systems must allow users to specify:

1. A series of an arbitrary number of consecutive chirps, enabling time dependent measurements.
2. Individual DC field offsets for each chirp within the series, enabling DC-field dependent measurements.
3. Individual Chirp Amplitudes for each chirp within the series, enabling AC-field dependent measurements.
4. The harmonic of interest for each chirp within the series, enabling investigation of higher order harmonics

Here, spectroscopic measurements are enabled by allowing the user specify each of the above parameters in any order or combination. This near-complete control of measurement specification not only enables all of the common measurements (e.g. field-dependent switching spectroscopy, field- and harmonic- dependent electromechanical non-lineariy measurements, and time- and field-dependent electromechanical relaxation measurements), but also streamlines development of alternative measurement schemes. Note that, *excitation waveform* (e.g. Fig. 2.2) is used to refer to the chirp and *measurement waveform* (e.g. Fig. 4.2) is used to refer to the user-specified series of chirps, DC- and AC- offsets, etc that represent a specific measurement scheme (e.g. a switching spectroscopy measurement).

2.2 Collection Path

Once the measurement waveform is applied to the material, all that remains is to capture the material response (Fig. 2.1 path b). First, the raw AC deflections are captured as a time series at the sampling rate specified by the user. With the knowledge of the exact measurement waveform, the deflections due to individual chirps can be extracted and transformed to the frequency domain via a fast Fourier transform (FFT). Then, the effects of the chirp are removed from the frequency response by deconvolution, yielding the material's frequency response (illustrated in Fig. 2.1 path b). A functional fitting of the frequency response to a damped driven oscillator (Eq. 1.1) can then used to extract the response parameters (amplitude A, phase ϕ, resonant frequency ω_0, and quality factor Q). Note that as discussed in the previous section, the user can specify which harmonic the fitting is applied to. Finally, the initial measurement waveform is again consulted to determine the conditions under which individual chirps were applied (e.g. time step, DC-field, and AC-field), allowing the resulting parameters (A, ϕ, ω_0, and Q) to be investigated as a function of these conditions.

2.3 Requirements

In its simplest form a RPFM systems only requires a means of application and collection as described above. However, here we impose two additional constraints onto our RPFM system. First and foremost, the RPFM system must be widely accessible, implemented on readily available hardware and applicable to a wide range of AFMs. To maintain applicability to most AFMs, an independent and standalone RPFM system is desired that requires minimal communication with the AFM. Second, the user must have complete control over the measurement waveform. As discussed, some RPFM systems impose arbitrary limits on acquisition time. Here, we seek to develop a system that is only limited by physical constrains (e.g. the Nyquist limit).

Thus, the RPFM system developed here must:

1. Generate a tapered linear swept-frequency sinusoid (or chirp) according to user specified parameters.

2. Allow for the chirp to be overlaid a user specified arbitrary waveform with user specifiable parameters to include: DC-field, AC-field, and harmonic of interest.

3. Acquire the raw response from the applied waveform and extract the material response.

4. Apply a functional fitting to the material response, extracting amplitude A, phase ϕ, resonant frequency ω_0, and quality factor Q.

5. Be implemented on commercially available hardware.

6. Require limited interaction with the AFM.

CHAPTER 3
DESCRIPTION OF EXCITE

3.1 Introduction

RPFM is a stand-alone system intended to be used in conjunction with an arbitrary AFM and is implemented through a combination of National Instruments hardware and custom Labview-Python software. RPFM allows for complete specification of a tapered linear chirp and allows users to overlay this chirp onto an arbitrary waveform. This measurement waveform can then be used to probe piezoresponse with any AFM that provides an electrical path to the tip and makes the raw AC tip deflections accessible. Further, the only hardware necessary for RPFM is readily available from National Instruments and the developed software is completely open-source. In the subsequent sections, the hardware and software are described.

3.1.1 AFM-Agnosticism

In order to ensure applicability to a wide range of AFMs, RPFM's communication with the AFM is minimized. This constraint results in RPFM being a stand-alone wide-band signal generation and data acquisition system that is essentially applicable to any tool.

In order to maintain an AFM-agnostic system, RPFM limits the communication with the AFM to only the absolute necessities. Communication between the system and an AFM must, at a minimum, consist of two signals. First, the measurement waveform generated by the RPFM system must be passed to the AFM and, second, the AFM must provide the raw tip deflections for the RPFM system to process. However, the RPFM system must also be informed *when* the measurement waveform should be applied and raw tip deflections acquired and, thus, communication cannot be limited to these 2 signals. To circumvent this timing issues, a third trigger signal must be used as well. The trigger signal is simply a rising edge signifying that RPFM should begin outputting the measurement waveform and acquiring the raw AC deflections. For each rising edge, RPFM will output the measurement waveform and acquire the response to the waveform only once. This single acquisition is referred to as an *Acquisition*.

An important consequence of this AFM-agnosticism is it's lack of knowledge about the tip movement. RPFM simply provides the specified measurement waveform and acquires the raw response whenever the AFM requests, i.e. whenever the AFM sends the trigger signal. This means that the user must be careful to keep track of how the measurement was performed on the AFM, so that results can be properly interpreted. Note that RPFM has a number of built in note taking tools (i.e. the Measurement Type, Measurement Name, and Plot Group fields) that are available to the user for this purpose.

3.1.2 The Measurement Waveform and Acquisitions

As discussed in the Theory section, the measurement waveform is an arbitrary and user-specified sequence of chirps, that represent some time- and field-dependent spectroscopic measurement (e.g. a switching spectroscopy measurement). Note again, that each time RPFM receives a trigger from the PFM, the measurement waveform is applied and the response acquired, with a single acquisition being referred to as an *Acquisition*. During operation of RPFM, the user provides the system with the number of acquisitions (i.e. number of triggers) that can be expected, and the system collects, processes, and stores the results in terms of these acquisitions. Thus, operation of RPFM is fundamentally based upon the measurement waveform.

3.1.3 A Brief Summary and Illustration

In summary, RPFM has a AFM-agnostic, trigger activated, and measurement-based scheme, which is best illustrated with a brief example. For instance, consider performing a point-by-point 10x10 grid based piezoresponse switching measurement (e.g, [23]). From RPFM's perspective, the user would specify a chirp, a switching measurement waveform to be overlaid the chirp, and $10 \times 10 = 100$ acquisitions. The system would then wait for 100 triggers from the AFM. Upon each trigger, RPFM will apply the switching measurement waveform and collect, process, and store the individual switching curves. This is all performed without any knowledge from RPFM about how the AFM tip is moving.

3.2 Hardware

3.2.1 Complete Hardware List

The physical components of Excite are: the National Instruments PXIe-1082, the National Instruments BNC-2110, and a computer monitor, keyboard, and mouse. The National Instruments (NI) PXIe-1082 (Fig. 3.1a) is a chassis that houses various modules. The PXIe-1082 contains 5 modules:

- PXIe-8880: An embedded controller that runs Windows 10. It is connected to the computer monitor, mouse, and keyboard.
- PXIe-5122: An oscilloscope/digitizer that is currently unused. It could possibly be used for data collection.
- HDD-8261: The hard drives for the system.
- PXIe-5412: A waveform generator that is currently unused. It could be used as an alternative means of waveform generation.
- PXIe-6124: A multifunction I/O module. This module is the workhorse of the RPFM system. It is used for waveform generation and collection. This module is connected to the BNC-2110.

The National Instruments (NI) BNC-2110 (Fig. 3.2) is connected to the PXIe-6124 multifunction I/O module and is used for signal input/output for the PXIe-6124.

a)


b)


Figure 3.1: a) Picture of the NI PXIe-1082. The arrows indicate the various connections. b) The monitor that is connected to the PXIe-8880 and is used to control the RPFM-PFM system.

3.2.2 Comments on Hardware

The hardware for RPFM only serves two purposes: generating the measurement waveform during the application path and acquiring the raw response in the collection path. Thus, the only necessary hardware is an arbitrary waveform generator, for application, and an analog-to-digital converter, for data collection. The NI PXIe-6124 and the connected BNC-2110 fulfills both of these roles. For the application path, the waveform is calculated in the software (described below) and is generated by the PXIe-6124. The generated waveform is output on one of the channels of the BNC-2110 (the default channel is AO 0, see Fig. 3.2) and the user accordingly directs the signal to the PFM tip (see, for instance, the procedure for the Cypher AFM). For the collection path, the analog-to-digital converter collects the raw AC deflections of the tip and all of the post processing to obtain the material response is achieved in the software. The analog-to-digital converter used here is an input channel of the PXIe-6124 (the default channel is AI 0, see Fig. 3.2). The user must direct the raw AC deflections of the tip to the PXIe-6124 to facilitate data collection.

Note that the PXIe-6124 has a maximum sample rate of 4 MHz, restricting the frequency range of the measurement waveform to < 2 MHz according to Nyquist-Shannon sampling theory [CITE], and a maximum voltage amplitude of ± 10 V. This limit on the voltage amplitude is an *absolute* limit and no point in the waveform can go beyond this limit. Thus, if the excitation chirp has an amplitude of 1.5 V then the maximum DC voltage that can be added to the chirp is 8.5 V.

As a final note on hardware, the above discussed trigger signal is provided to the PXIe-6124 through a trigger channel (the default channel is PFI 12, see Fig. 3.2). When a rising edge is applied to the trigger channel, the PXIe-6124 will send the measurement waveform, once and only once, on the selected output channel and will sample the response on the selected input channel.

In summary, for proper operation of RPFM, three signals must properly routed through the PXIe-6124: the measurement waveform, the trigger signal, and the response. The measurement waveform is the waveform that is generated by the PXIe-6124 and is applied to the material through the PFM tip. The trigger signal is what prompts the PXIe-6124 to output the probing waveform and begin data collection. The response is the acquired material response that the AFM outputs to the PXIe-6124. These three signals are communicated to/from the PXIe-6124 through the BNC-2110.

3.3 Comments on Nyquist-Shannon Sampling Theory

In brief, the Nyquist-Shannon Sampling theorem [CITE] states that when either generating or digitizing an analog signal the sample rate must be greater than twice the highest frequency in the signal. The upper limit on the sampling rate is typically a practical limitation imposed by either the hardware or memory usage, as higher sample rates will result in more sample points and, thus, more memory. For instance, consider a typical RPFM measurement with $f_L = 250\ kHz$ and $f_H = 300\ kHz$, here a sample rate of at least $600\ kHz$ necessary. However, the Nyquist limit is merely a minimum recommendation and using a sample rate of $600\ kHz$ will eliminate the possibility of probing the higher order harmonics. Thus, it is recommended to use a reasonably high sample rate. In this case, a sample rate of $2\ MHz$ will allow for the extraction of the fundamental, second, and third harmonic if desired.

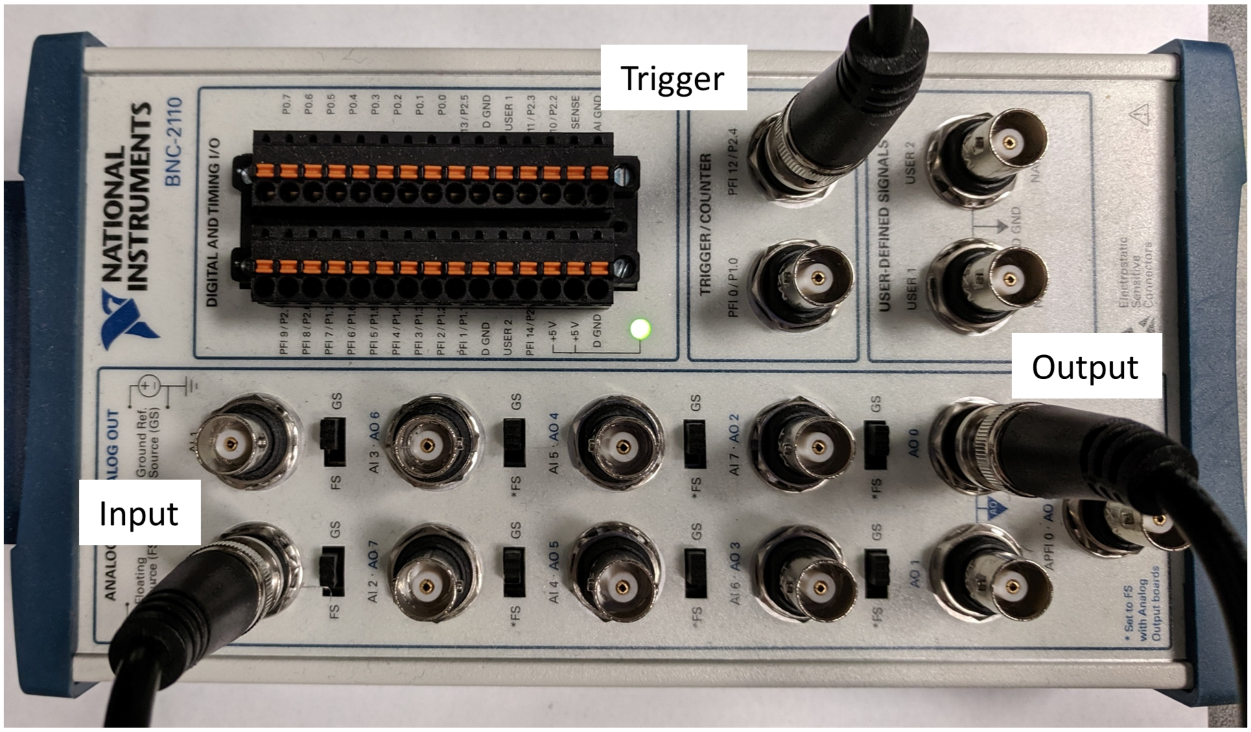

Figure 3.2: Picture of the NI BNC-2110. White boxes label the signal paths.

3.4 Software

3.4.1 Software

The software that runs RPFM is called Excite and is shown in its initial state in Fig. 3.3. This is the software that will run in concert with the PFM and handles: managing the PXIe-6124, calculating the measurement waveform, some initial post-processing, and more. This software is divided into two components the python app and the labview app. The python app collects the user inputs, passes them to the Labview, generates supplementary files, and performs some post processing as the raw data is received from the PXIe-6124. The Labview app receives the user's specifications from python, calculates the measurement waveform, manages the PXIe-6124, and saves the raw data in the specified folder. During typical operation of Excite, the user should only interface with the python app. The Labview app only needs to be used if the user wishes to change options related to the hardware (e.g. change the output port).

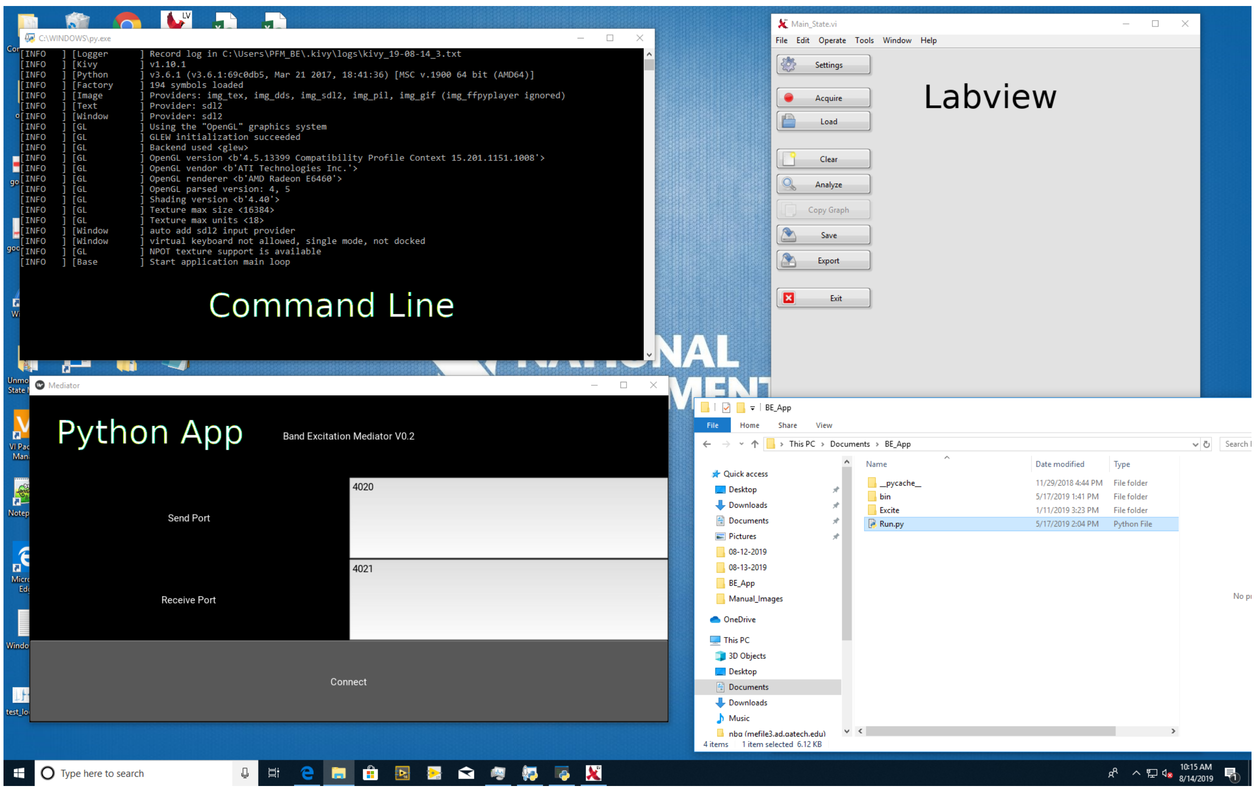

Figure 3.3: Excite software upon initally launching software. Large text within the various windows labels the which component of the software the window is from

CHAPTER 4

EXCITE OVERVIEW OF OPERATION

When operating Excite, there are three task: prepare the physical connections between the AFM and the BNC-2110, prepare the Excite software, and prepare the AFM. Here, we will walk-through all three for a generic AFM. This overview is not intended as strict instructions, but as a means of illustrating how Excite operates. For these specific instructions see Chapter 5, which describes how Excite is operated with an Asylum MFP-3D and Cypher.

4.1 Types of Measurements

For the purpose of discussion, we can roughly classify the type of measurements typically performed on a AFM into two categories: scans, which raster over the surface while probing the response and point-by-point measurements, which probe the response at a single point or at a series of individual points over time and typically in response to some additional applied field.

4.2 The Physical Connections

As discussed above, proper operation of Excite requires the appropriate routing of three signals. First, the measurement waveform (by default channel *AO 0* of the BNC-2110) must be routed to the AFM tip and, second, the raw AC deflections of the tip must be routed to the analog-to-digital converter (by default channel *AI 0* of the BNC-2110). These signals are covered in Chapter 3. The final signal is the trigger signal which should be routed from the AFM to the trigger channel of the PXIe-6124 (by default channel *PFI 12* of the BNC-2110).

4.3 Excite Software

4.3.1 Starting Excite Software and Connecting Python and Labview

To start Excite, double click *Run.py* located in the *1Acquisition* folder. This launches both the python app and the labview app and should bring up the three windows shown in Fig. 3.3. Prior to operation, the python and labview app must first be connected through TCP/IP. Connecting the two apps simply involves specifying the send and receive port numbers and clicking *Connect* in the python app. It is through this TCP/IP connection that the python app transmits the user inputs (send port) to Labview and the Labview app responds (receive port). The default port numbers are 4020 for the send port and 4021 for the receive port. Note, that the user should not change these port numbers without a basic understanding of networking and, if these numbers are changed, corresponding changes must be made within the Labview app (in the file INFONEEDED FILE) and the python app (in the file INFONEEDED FILE).

A successful connection is shown in Fig. 4.1. Note that the Command Line window now has 5 new lines: "$Sending\ connection\ message\ to$ 4020" and "$sent\ \{"message\_name";"connect"\}$" indicating that python sent a "connect" message to Labview, "$Connection\ from\ \{127.0.0.1,\ S0312\}$" and "$\{"message\_name";"connectAck"\}$" indicating that python received a connectAck (or connection acknowledgement) from Labview, and "Connected" indicating that the connection has been established. Also, note that the python app has updated to the parameter specification screen. Now that the python and Labview apps are connected, the user can begin to input the measurement specifications.

Troubleshooting

- *Failure to connect:* When attempting to connect, if the command line shows that python sent a connect message but shows that no acknowledgement message was received, then the connection has failed. The user should check: first, that the correct port numbers were used and, second, that only one instance of each app is open.

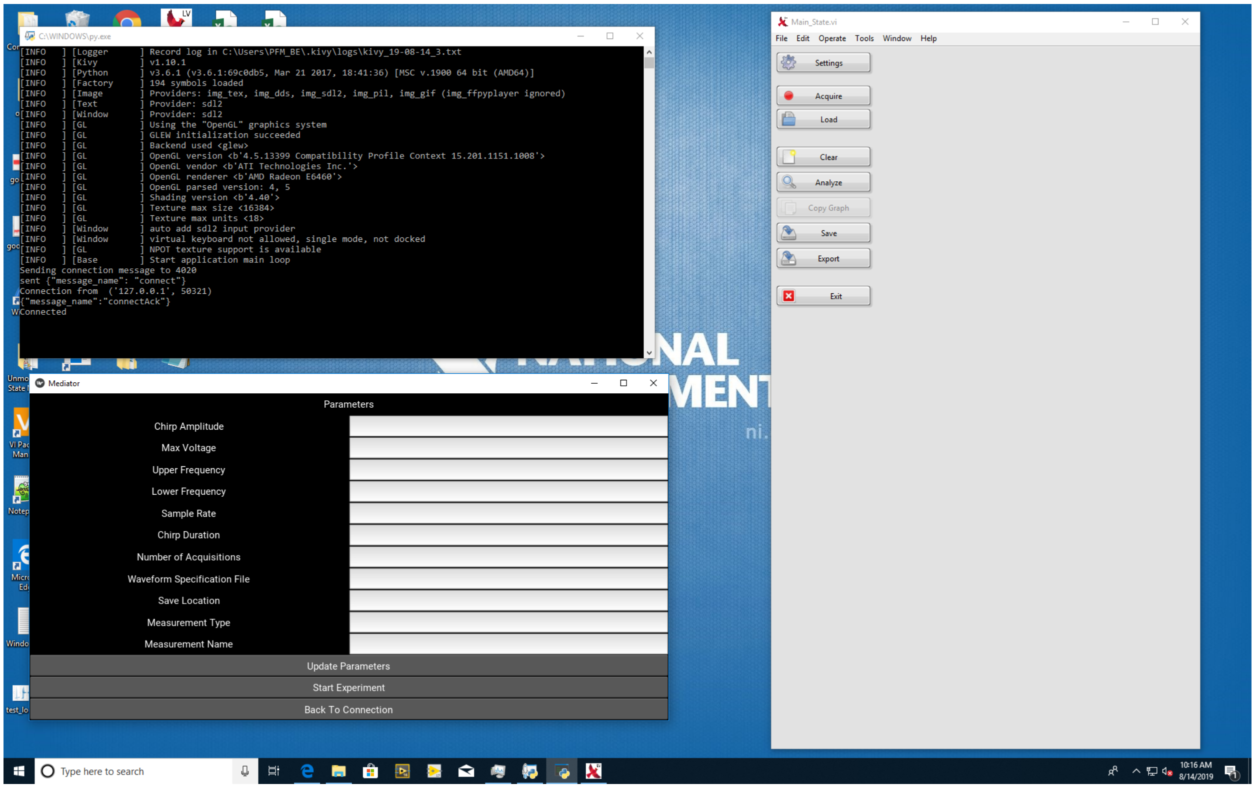

Figure 4.1: Excite software upon initially launching software. Large text within the various windows labels the which component of the software the window is from

4.3.2 Specifying the Excitation Waveform

First, the user should specify the excitation waveform (i.e. the chirp). The parameters specified by the user in python are the Chirp Amplitude, Lower Frequency, Upper Frequency, Sampling Rate, and Chirp Duration. These parameters are input to the python app, transmitted to Labview over TCP/IP, and used to calculate the excitation waveform in Labview according to Eqs. 2.1 and 2.2. Appropriate values for Chirp Amplitude, Lower Frequency, and Upper Frequency are strongly dependent on the sample and PFM tip in question. It is best to empirically obtain these values by performing some initial frequency sweeps, such sweeps can be performed in via a lock-in sweep typically available in any single frequency PFM mode.

A note regarding Chirp Amplitude: If a voltage amplifier external to the PXIe-6124 and BNC-2110 is being used, the user should account for the amplification here. For example, if the waveform being output by the PXIe-6124 is being amplified by a 10X voltage amplifier and the user desires a 1.5 V chirp, then the user must specify a chirp amplitude of 0.15 V in Python.

4.3.3 Specifying the Measurement waveform

Next, the user can specify a measurement waveform. As discussed in Band Excitation Theory in Chapter 2, the excitation waveform can be overlaid an arbitrary waveform, enabling voltage- and time- dependent spectroscopic measurements. This arbitrary waveform is specified in a waveform specification file (WSF). The WSF is a comma separated values (.csv) file with a specific structure, consisting of an arbitrary number of rows and 5 columns. From left to right, the columns in the WSF correspond to *Duration*, *DC Offset*, *Multiplier*, *Harmonic*, and *Plot Group*. A path to this file is input in the python app at the Waveform Specification File entry and python passes this path to the Labview app. Labview then reads the WSF row by row, from top to bottom. At each row, Labview constructs a series of excitation chirps based off the *Duration*, *DC Offset*, *Multiplier*, *Harmonic*, and *Plot Group* for that row, appending the chirp series together as it works down the rows. A description of what the individual columns specify follows.

- *Duration:* The *Duration* is the number of consecutive chirps in the series for the row and must be an integer. For example, a *Duration* of 3 would result in a series of three consecutive chirps.

- *DC Offset:* The *DC voltage* value is a constant value added to the chirp series for this row, and is specified in volts. Continuing the previous example, a *Duration* of 3 and a *DC Offset* of 1 would result in a series of three consecutive chirps with a 1V DC offset added.

- *Multiplier:* The *Multiplier* is an arbitrary value to multiply the excitation chirp series by for this row, and is unit-less. This is the parameter that the user can use to vary the AC-Field (or Chirp Amplitude) throughout the measurement waveform. This value **DOES NOT** affect the DC offset, just the amplitude of the chirps. For instance, a Chirp Amplitude of 2V, a *Duration* of 3, a *DC Offset* of 1, and a *Multiplier* of 2 would result in a series of three consecutive chirps with a Chirp Amplitude of 4V and with a 1V DC offset added.

- *Harmonic:* The *Harmonic* option can be either 1, 2, or 3 and specifies which harmonic of the frequency response is of interest for this row. This option is not actually used in the generation of the measurement waveform, but specifies over which frequency range the functional fitting to Eq. 1.1 should be performed. For instance, consider if the chirp frequency parameters are: $f_L = 250\ kHz$ and $f_H = 300\ kHz$. A *Harmonic* value of 1 for a row would result in the functional fitting being applied over the frequency range $250\ kHz$ to $300\ kHz$, while a harmonic value of 2 would result in the fitting being applied over $500\ kHz$ to $600\ kHz$. Note, be sure to use and appropriate sample rate. In the example here, at least $2 \times 600\ kHz = 1.2\ MHz$ would be required.

- *Plot Group:* The *Plot Group* is an arbitrary identifier for this row, provided for the user. This option is never used in the measurement waveform generation or during fitting. However, the final results will retain their associated *Plot Group* value and the user is welcome to use this option to allow for easy extraction of a particular subset of results. Note, that *Plot Group* can take any numerical value outside of 9 which is reserved.

Understanding the WSF files is best done through examples. Thus, a few examples follow. For these examples, we will only consider the Duration and DC Offset columns. First, an illustration of the proper WSF format is provided in Table 4.1 for the simplest measurement waveform, a single unaltered excitation chirp. As discussed, Labview constructs the measurement waveform by reading the WSF row by row, thus consider the first (and only) row in Table 4.1. A duration of 1 indicates that single chirp is desired for this row and a DC Offset of zero corresponds to no additional bias being added to the chirp. Thus, this WSF specifies a single excitation chirp with no modifications (Fig. 2.2) .

In Table 4.2, a WSF for a simple relaxation measurement is illustrated. Beginning with the first row, a duration of 5 indicates a series of 5 excitation chirps and a DC offset of 1 indicates that the 5 excitation chirp will be offset by a DC value of 1 V. In the next row, a duration of 10 indicates a series of 10 excitation chirps and a DC offset of 0 indicates no offset by a DC value. These two rows are generated consecutively. Thus, in total, the measurement waveform will consist

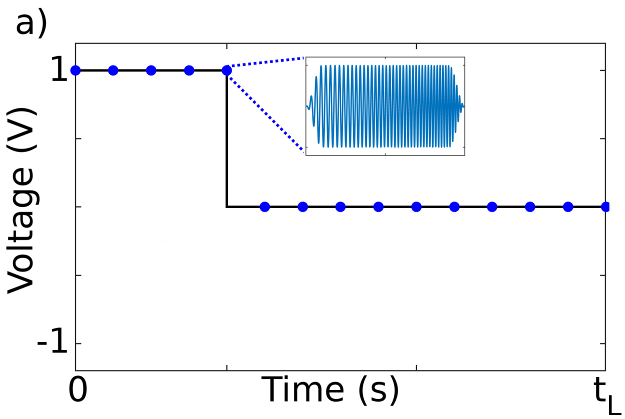

Figure 4.2: Illustrations of a) a relaxation measurement waveform specified by Table 4.3.3 and b) a switching spectroscopy measurement waveform specified by Table TABLE. a) A simple relaxation measurement waveform where five consecutive chirps are applied with an added DC voltage of 1V and a subsequent 10 consecutive chirps are applied with no DC bias. In this way, the material is probed for five points while being poled by 1V and the resulting electromechanical relaxation upon removal of the 1V is probed for 10 points. b) A simple switching spectroscopy measurement waveform where the materials is probed from -3V to 3V in the typical SS-PFM fashion. Blue dots represent a single chirp and the black line represents the DC voltage.

Duration	DC Offset	Multiplier	Harmonic	Plot Group
1	0	1	1	0

Table 4.1: Simple example of a waveform specification file. A file like this would specify a single chirp with no alterations.

of 5 consecutive chirps of probing under a 1V bias (in-field), followed by 10 consecutive chirps of probing under no bias (out-of-field). This waveform is plotted in Fig. 4.2a, where blue dots represent the individual chirps and the black line represents the DC voltage.

Duration	DC Offset	Multiplier	Harmonic	Plot Group
5	1	1	1	0
10	0	1	1	0

Table 4.2: Simple example of a relaxation waveform specification file. A file like this would specify 5 chirps under a 1V bias, followed by 30 chirps under no bias. This waveform is illustrated in Fig. INFO FIGURE.

In Table 4.3, a WSF for a simple SS-PFM measurement is illustrated. First, note that each row

Duration	DC Offset	Multiplier	Harmonic	Plot Group
1	1	1	1	0
1	0	1	1	0
1	2	1	1	0
1	0	1	1	0
1	3	1	1	0
1	0	1	1	0
1	2	1	1	0
1	0	1	1	0
1	1	1	1	0
1	0	1	1	0
1	-1	1	1	1
1	0	1	1	1
1	-2	1	1	1
1	0	1	1	1
1	-3	1	1	1
1	0	1	1	1
1	-2	1	1	1
1	0	1	1	1
1	-1	1	1	1
1	0	1	1	1

Table 4.3: Simple example of a PFM-based switching spectroscopy waveform specification file. A file like this would specify a typical -3V to 3V SS-PFM measurement and probed the response by in-field (during the applied DC voltage) and out-of-field (upon removal of the DC voltage).

has a Duration of only 1 and, thus, each row only represents a single chirp. Further, the DC Offset column has the structure of a typical SS-PFM measurement ranging from -3 to $3\ V$. That is to say, the DC Offsets show a positive voltage triangle, followed by a negative voltage triangle, with zero DC field points in between each step. This waveform is illustrated in Fig. 4.2b.

4.3.4 Number of Acquisitions

Once the user has specified the excitation and measurement waveform, they must next specify the number of *Acquisitions*. As stated above, an *Acquisition* is a single application of the measurement waveform and subsequent acquisition of the material's response and is triggered anytime a rising edge is applied to the trigger channel of the PXIe-6124. In the Python app, the user must specify the number of *Acquisitions*, or the number of triggers, that Excite should expect. For instance, if a user wanted to apply some arbitrary measurement waveform (e.g. an SS-PFM waveform, Fig. 4.2b) at 100 points on a surface area, then *Number of Acquisitions* must be specified as 100. Consider another example, typical PFM scan has a resolution of 256 by 256 points and is acquired by scanning the PFM tip over the surface area collecting 256 point per line for 256 lines (illustrated in FIG). To incorporate into this measurement is the user has two options: specify *Number of Acquisitions* as $256 \times 256 = 65536$ and provide a trigger signal for each point in the 256 by 256 grid or specify *Number of Acquisitions* as 256, use a WSF to specify a train of 256 chirps (Table 4.3.4, and provide a trigger signal for each line. Note that the latter is the recommended approach.

Duration	DC Offset	Multiplier	Harmonic	Plot Group
256	0	1	1	0

Table 4.4: Waveform specification file for capturing a single line of a PFM scan with 256 points per line.

4.3.5 Remaining Measurement Parameters

The remaining measurement parameters are: Max Voltage, Save Location, Measurement Type, and Measurement Name. Max voltage is the voltage limit on the measurement and is the maximum allowable voltage output of the PXIe-6124. Note that the hardware limits the voltage out of the PXIe-6124 to $\pm 10\ V$. However, with this option, the user can impose a software limit if desired. Save Location is the folder in which all of the raw data, processed data, and supplementary files will be saved. Measurement Type and Measurement Name are only used by the tool bepy, which is a supplemental tool for plotting, data organization, and analysis. However, neither of these parameters is necessary for Excite to operate properly. Measurement Type is a tag used by the tool bepy and should be set to either Scan, Relax, SSPFM or NonLin, which correspond to a basic RPFM domain imaging measurement, a relaxation measurement, a switching spectroscopy measurements, and a non-linear measurement. Naturally, when performing one of these four measurements the

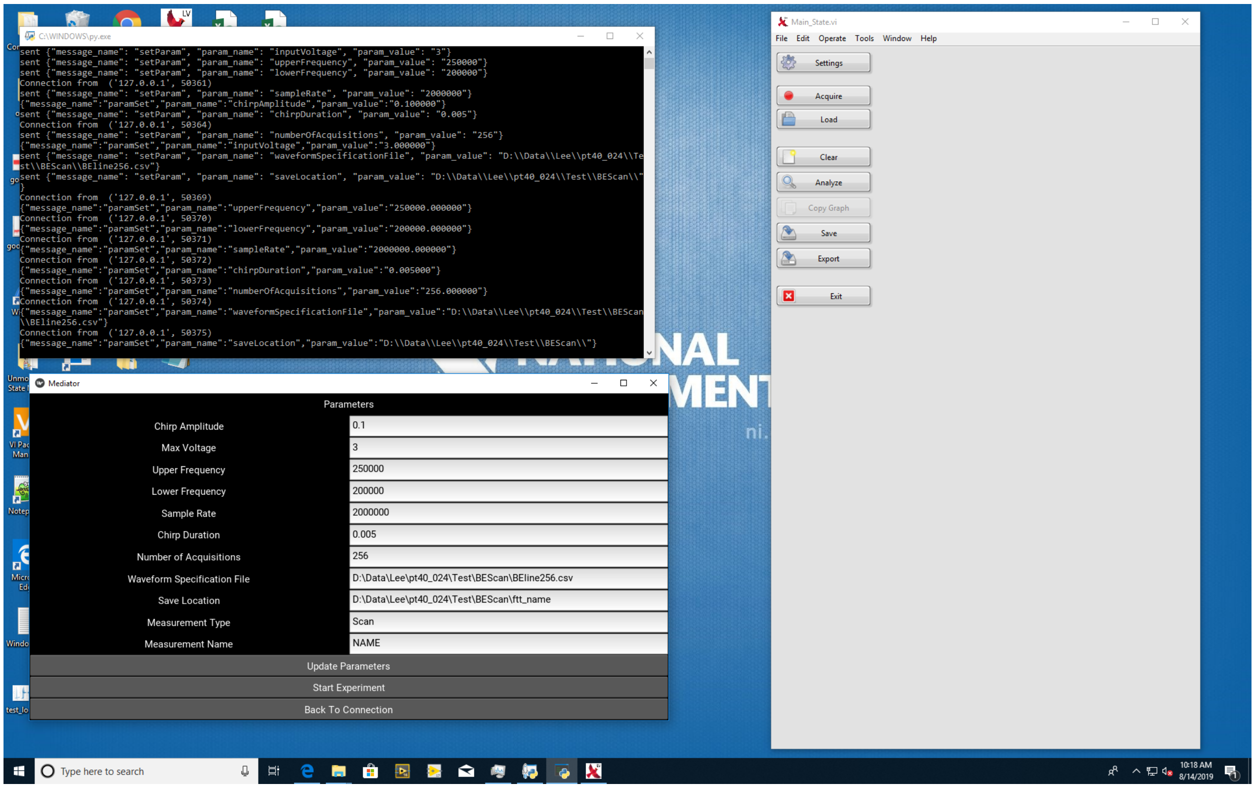

Figure 4.3: Excite software with arbitrary parameters and after clicking *Update Parameters*.

corresponding tag should be used for Measurement Type. However, if the measurement is none of these, one of these four tags must still be chosen. In this case, the user should choose whichever tag is appropriate for the measurement. Note that bepy by default plots Scan-types as an image of the individual *Acquisitions*, Relax-types as a function of time, SSPFM-types as a function of the DC Offset in the WSF, and the NonLin-types as a function of the Multiplier in the WSF. Finally, the Measurement Name parameter is used by bepy as a unique identifier for the measurement and can be any string of characters, numbers, and/or special characters.

4.3.6 Confirming Measurement Parameters

Once all of the above parameters have been specified, the user must click *Update Parameters* which sends all of the necessary parameters from the Python app to the Labview app. At this point the Excite software should resemble Fig. 4.3. Note that the Command Line window now has numerous new lines consisting of: "$Connection\ from\ \{127.0.0.1,\ S0312\}$" indicating the connection, sent

$\{''message\_name'' : ''setParam'', ''param\_name'' : ''paramIdentifier'', ''param\_value'' : ''3''\}$ indicating that Python has told Labview to update the parameter named "paramIdentifier" to the some value (here "3"), and $\{''message\_name'' : ''paramSet'', ''param\_name'' : ''paramIdentifier'', ''para$ $''3''\}$ indicating that Labview has updated the parameter named "paramIdentifier" to the requested value.

4.3.7 Summary

Once all of the parameters have been specified and confirmed, the Excite software is ready to begin the specified measurement. At this point, clicking *Start Experiment* will cause Excite to begin listening for triggers from the AFM. However, it is recommended that the user prepare the AFM before clicking *Start Experiment*, as Excite will time-out if a trigger isn't received within 2 minutes.

4.4 Preparing the AFM

4.4.1 Syncing

To reiterate, a side effect of Excite's AFM-agnosticism is that Excite has no control or knowledge of the tip movements. Thus, it falls to the user to ensure that the Cypher is properly "synced" with Excite. In other words, when the Cypher provides a trigger to Excite to begin application and acquisition, the user must ensure that the Cypher provides adequate time for the application and acquisition to finish. In a scan measurement, this typically means specifying an appropriate scan rate and accounting for any overscan the AFM might do automatically when scanning. In a point-by-point measurement, this typically means ensuring that the tip dwells long enough at the surface for the application and acquisition to finish. The specifics of syncing will depend on the particular AFM used and, thus, will be more completely discussed in Chapter 5 for the Cypher and MFP3d.

CHAPTER 5

EXCITE OPERATION MANUAL

Note: The directions given below assume that the default channels on the BNC-2110 are selected (AO 0 is outputting the measurement waveform, AI 0 is acquiring the response, and PFI 12 is the trigger channel). Further, note that any of the BNCs on the Cypher are usable, but in these instructions particular ones are recommend for ease of use.

5.1 Comments on Preliminary PFM

Preliminary basic PFM tunes and scans are highly recommended. In general, best practice dictates first using basic PFM to: find a suitable area, perform numerous tunes, perform (at a minimum) a single vertical and lateral scan, and perform read/write or simple switching spectroscopy in the surrounding area. The following instructions for applying Excite assumes the user has performed all or some of these steps and has a good idea of: the coercive voltages of the sample area and the expected resonant frequency.

5.2 Excite on the Cypher

5.2.1 Comments on Cypher Operation

Crosspoint Panel

The Cypher has numerous BNC connections available to the user capable for both input and output. Further, the software provided by Asylum allows the user to control how these BNC signals are routed throughout the Cypher, this is achieved through the **Crosspoint Panel** (shown in Fig. X). The **Crosspoint Panel** is opened by going to the Programming tab in the Asylum software and clicking ”Crosspoint Panel”. The crosspoint panel is divided into three columns. The left column corresponds to signals and BNC connections in/on the Cypher itself. The right column corresponds to signals and BNC connections in/on the ARC2 SPM controller. The middle is other signals. Here, we use only the left column (the signals/connections on the Cypher), although other schemes are possible.

For operation of Excite, the most important options are:

- **PFMHV**: specifies the signal is input to the internal 10x voltage amplitfier and routed to the AFM tip.
- **BNCOut0, BNCOut1, etc.**: specifies what signals should be output on the available BNC output channels.

Finally, note that the ”lock” icon next to changed options should be clicked (the locks will become red), otherwise the software will likely change them back. Then ”Write Crosspoint” should be clicked once all options have been changed.

The ARC2 SPM Controller, the Trigger Signals, and the Trigger Control Box

The ARC2 SPM controller controls the Cypher and most of the Asylum SPM's. The ARC2 provides three trigger signals:

- A *point trigger*: signifies the beginning of a point
- A *line trigger*: signifies the beginning and ending of a line
- A *frame trigger*: signifies the beginning and ending of a frame

These signals are provided by the Expansion (Fig Y) connection on the front of the ARC2. Specifically, the *point trigger* is on pin 17, the *line trigger* is on pin 4, and the *frame trigger* is on pin 16.

A Trigger Control Box (red 3d printed box) (Fig Z) was developed to extract the *point trigger* and the *line trigger* and provide a "thru" connection so that other expansions can still be plugged into the ARC2. One side provides the point and line triggers as separate BNC connections. The other side has a *Trigger* and a *Control* BNC connection. Here, the *Trigger* connection provides the *point trigger* when no voltage is applied to *Control*. When 5V is applied to *Control*, the *Trigger* connection provides the *line trigger*.

5.2.2 Cypher Scan Measurements with Excite

Preparing Physical Connections

WARNING!!!!!! NEVER CONNECT AN INPUT TO AN INPUT OR AN OUTPUT TO AN OUTPUT

1. Connect the output channel on the BNC-2110 (**AO 0**) to an input BNC on the Cypher (**BNCIn0**)

2. Connect an output BNC on the Cypher (**BNCOut0**) to the acquisition channel on the BNC-2110 (**AI 0**).

3. Connect the trigger channel on the BNC-2110 (**PFI 12**) to *Line* on the Trigger Control Box

    - Or: Connect the trigger channel on the BNC-2110 (PFI 12) to the *Trigger* on the Trigger Control Box.
    - Connect the *Control* on the Trigger Control Box to BNCOut1.
    - Open the Crosspoint Panel and set BNCOut1 to 5VRef. Which applies 5V to the *Control* channel on the Trigger Control Box, setting the *Trigger* channel to *Line*.

Notice: Here we are using the *line trigger* and, thus, Excite will be begin application of the measurement waveform and acquisition at the beginning of each line! The waveform specification file (WSF) MUST reflect this!

Preparing the Excite Software

1. Run 'Run.py'

2. Connect the Python and Labview app. (See Chapter 4 for details)

3. Specify (See Chapter 4 for details):

   - Chirp Amplitude: Typically 0.5V to 1.5V. HOWEVER, the Cypher contains an internal 10x amplifier. Thus, choose 0.05V to 0.15V.
   - Max Voltage: Typically 1-3V
   - Upper Frequency: Based upon the preliminary scans and tunings, choose a range that will best capture the resonant peak. Typical bandwidths are 40-50 kHz. Typical Upper Frequencies range from 250 kHz to 450 kHz.
   - Lower Frequency: Based upon the preliminary scans and tunings, choose a range that will best capture the resonant peak. Typical bandwidths are 40-50 kHz. Typical Lower Frequencies range from 210 kHz to 410 kHz.
   - Sample Rate: The max available Sample Rate is 4 MHz. At this point, the user should consider Nyquist Sampling Theory (discussed in Chapters 2 and 3). Often 2 MHz is suitable.
   - Chirp Duration: This should be set to 0.006 s. Other values are acceptable, but the user should calculate a suitable scan rate for the new Chirp Duration.
   - Number of Acquisitions: Choose 256 for a 256 by 256 scan.
   - Waveform Specification File: Provide a path to the .csv waveform specifical file. (Including the name of the file)
   - Save Location: Typically, choose the same location as the Waveform Specification File. Although any path is acceptable. Be sure to provide a file name at the end of the path. E.g. C:\Documents\Data\my_save_name

- Measurement Type: Type 'Scan'
- Measurement Name: Any string is acceptable. Choose something that identifies this measurement.

4. The recommend WSF for a Cypher scan is shown if Table 5.1. The reasoning for this choice is discussed below.
5. Click 'Update Parameters'

Notice: Continuing with this "line based" scan scheme, we select number of acquisitions as 256! This is because we want 256 lines in a 256 by 256 scan! Further, note that the WSF in Table 5.1 is set up to send 256 chirps (plus a few additional, which is discussed below) each time it is triggered, i.e. for each line! This results in the desired 256 by 256 scan image.

Duration	DC Offset	Multiplier	Harmonic	Plot Group
19	0	1	1	0
256	0	1	1	1
19	0	1	1	0

Table 5.1: Waveform specification file for capturing a single line of a PFM scan with 256 points per line.

Preparing the Cypher - Crosspoint

1. Open the **Crosspoint Panel**.
2. Set **PFMHV** to the input BNC (BNCIn0). Click the lock next to **PFMHV**.
3. Set the output BNC (**BNCOut0**) to ACDefl. I.e. the raw AC deflections of the tip. Click the lock next to **BNCOut0**.
4. Click "Write Crosspoint" on the **Crosspoint Panel**.

Preparing the Cypher - Syncing

Notice: By default, scanning in the Cypher scans each line twice (a trace and a re-trace). Additionally, the Cypher "overscans" the area by about 7.5% on the left and the right. This overscan is present to account for the tip reversing direction at the end of each line. In this example, we seek to capture a 256 by 256 trace scan with Excite. First, we begin with the WSF show in Table 5.1. Here, 256 chirps are flanked by 19, about 7.5% of 256, which accounts for the overscan. Now that the overscan has been accounted for, a proper scan rate should be calculated. The WSF in Table 5.1 has a total of $N_c = 256 + 19 + 19 = 294$ chirps and we have selected a Chirp Duration of $\Delta t = 0.006$. This yields $t_{trace} = \Delta t N_c = 1.764\ s$ per trace, but accounting for the re-trace this becomes $t_{line} = 2t_{trace} = 3.528\ s$ per line. This is a scan rate of $SR = \frac{1}{t_{line}} = \frac{1}{3.528} \approx 0.28$ lines per second.

1. Prepare a typical scan in the Asylum Software.

2. In the Asylum Software, set scan rate to 0.28 Hz.

Begin Measurement

Notice: Always start Excite FIRST, as Excite is the "listener", or waits for the AFM to send a trigger.

1. In the Excite software, click Being Experiment.

2. In the Asylum software, click Frame Down.

5.2.3 Cypher Point-by-Point Measurements with Excite

Preparing Physical Connections

WARNING!!!!!! NEVER CONNECT AN INPUT TO AND INPUT OR AN OUTPUT TO AN OUTPUT

1. Connect the output channel on the BNC-2110 (**AO 0**) to an input BNC on the Cypher (**BNCIn0**)
2. Connect an output BNC on the Cypher (**BNCOut0**) to the acquisition channel on the BNC-2110 (**AI 0**).
3. Connect the trigger channel on the BNC-2110 (**PFI 12**) to *Point* on the Trigger Control Box
   - Or: Connect the trigger channel on the BNC-2110 (PFI 12) to the *Trigger* on the Trigger Control Box.
   - Connect the *Control* on the Trigger Control Box to BNCOut1.
   - Open the Crosspoint Panel and set BNCOut1 to Ground. Which applies Ground to the *Control* channel on the Trigger Control Box, setting the *Trigger* channel to *Point*.

Notice: Here we are using the *point trigger* and, thus, Excite will be begin application of the measurement waveform and acquisition at the beginning of each point! The waveform specification file (WSF) MUST reflect this!

Preparing the Excite Software

1. Run 'Run.py'
2. Connect the Python and Labview app. (See Chapter 4 for details)
3. Specify (See Chapter 4 for details):

- Chirp Amplitude: Typically 0.5V to 1.5V. HOWEVER, the Cypher contains an internal 10x amplifier. *Thus, choose 0.05V to 0.15V.*
- Max Voltage: Typically 1-3V
- Upper Frequency: Based upon the preliminary scans and tunings, choose a range that will best capture the resonant peak. Typical bandwidths are 40-50 kHz. Typical Upper Frequencies range from 250 kHz to 450 kHz.
- Lower Frequency: Based upon the preliminary scans and tunings, choose a range that will best capture the resonant peak. Typical bandwidths are 40-50 kHz. Typical Upper Frequencies range from 210 kHz to 410 kHz.
- Sample Rate: The max available Sample Rate is 4 MHz. At this point, the user should consider Nyquist Sampling Theory (discussed in Chapters 2 and 3). Often 2 MHz is suitable.
- Chirp Duration: This can be set to 0.005 s or greater
- Number of Acquisitions: Choose 1 for a single point. Choose $N \times M$ for a N by M grid, e.g. for a 50 by 50 grid choose 2500.
- Waveform Specification File: Provide a path to the .csv waveform specifical file. (Including the name of the file)
- Save Location: Typically, choose the same location as the Waveform Specification File. Although any path is acceptable. Be sure to provide a file name at the end of the path. E.g. C:\Documents\Data\my_save_name
- Measurement Type: Type 'SSPFM' for switching spectroscopy measurement. Type 'Relax' for a relaxation measurement. Type 'NonLin' for a non-linear measurement.
- Measurement Name: Any string is acceptable. Choose something that identifies this measurement.

4. For point-by-point measurements, the choice of WSF reflects the type of spectroscopy being performed. A rudimentary switching spectroscopy WSF is shown in (Table 4.3) and

relaxation WSF is shown in Table 4.2. In this example, we use the WSF for a rudimentary relaxation measurement. However, as will be discussed below, 10% of the total number of chirps is added at the beginning of the waveform and these chirps are set to zero amplitude by setting multiplier to zero.

5. Click 'Update Parameters'

Duration	DC Offset	Multiplier	Harmonic	Plot Group
12	0	0	1	9
20	1	1	1	0
100	0	1	1	0

Table 5.2: Simple example of a relaxation waveform specification file. A file like this would specify 5 chirps under a 1V bias, followed by 30 chirps under no bias. This waveform is illustrated in Fig. INFO FIGURE.

Preparing the Cypher - Crosspoint

1. Open the **Crosspoint Panel**.

2. Set **PFMHV** to the input BNC (BNCIn0). Click the lock next to **PFMHV**.

3. Set the output BNC (**BNCOut0**) to ACDefl. I.e. the raw AC deflections of the tip. Click the lock next to **BNCOut0**.

4. Click "Write Crosspoint" on the **Crosspoint Panel**.

Preparing the Cypher - Syncing

Notice: The Asylum software Do IV Panel (Fig N) allows for the specification of an arbitrary waveform by selecting the 'Function Editor' for Function. Here, we can specify the time length of the waveform being applied to the surface, a convenient means of syncing Excite and the Cypher. However, it is advisable to include a small amount of "settle time" to account for any latency in the process. In other words, a small amount ”blank chirps” should be included at the beginning of the measurement waveform to ensure that the tip has come in good contact with the surface. The recommended settle time is approximately 10% of the total number of chirps in the waveform. For instance, in Table 5.3, 12 chirps with a multiplier of zero (i.e. zero amplitude chirps) are added to the front of a 120 chirp measurement waveform, bringing the total number of chirps to $N_c = 132$. Next, another 10% of the original number of chirps should be added to the total without adding them to the WSF, thus $N_c = 154$. This adds more room for error, which is necessary for maintaining the synchronization between the Cypher and Excite. Given that $\Delta t = 0.005$, the required time for application of the measurement waveform and acquisition can be found with $t_t = \Delta t N_c = 0.77\ s$.

1. Prepare a typical single point force or force map in the Asylum Software. The recommended trigger point is a ”Relative” 2V Deflection.

2. In the Asylum Software Do IV Panel, change the Function to ’Function Editor’.

3. Set the length of the Function to $0.77\ s$.

Begin Measurement

Notice: Always start Excite FIRST, as Excite is the ”listener”, or waits for the AFM to send a trigger.

1. In the Excite software, click Being Experiment.

2. In the Asylum software, click Frame Down or Single Point Force.

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