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A temperature oscillation instrument to determine pyroelectric properties of materials at low frequencies: Towards elimination of lock-in methods

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Pyroelectric properties of materials can be accurately determined by applying a new digital signal processing method on the discrete sampled data obtained with a temperature oscillation technique. The pyroelectric coefficient is calculated from the component of the generated current 90° out of phase with respect to the sinusoidal temperature wave. The novelty of the proposed approach lies in the signal analysis procedure which implements a simple Fast Fourier transform that filters residual noise through convolution, and calculates the phase difference between the peaks of the temperature and current waves. The new idea requires relatively simple hardware and enables very accurate measurement of the pyroelectric coefficient of materials at ultra low frequencies, 1–250 mHz, without using costly lock-in amplifiers. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4932678>]

I. INTRODUCTION

Pyroelectric materials show a temperature dependent spontaneous polarization, as a result of the presence of a bound charge layer that attracts electrons and ions on the electrodes perpendicular to the polar axis of the material. If such a material is connected to an ammeter with a low internal resistance, there would be no current flow since the polarization is constant. A change in the temperature of the pyroelectric material, accompanied by a change in the net dipole moment, results in a displacement current across the electrodes perpendicular to the polar axis of the material.¹ The pyroelectric coefficient can be subsequently defined based on the generated pyroelectric current.

There are four main approaches in the literature for measuring the pyroelectric constant of the materials. (1) The charge integration method or direct measurement of the polarization, also known as the static method based on the integration of charge developed on the crystal faces as the temperature increases. This method is restricted to ferroelectrics and results are affected by ohmic conductivity and trapped charges in the bulk material and at the sample-contact interface.^{2–4} (2) The radiant heating or pulse method also known as the dynamic method,⁵ in which the sample is continuously heated by a modulated light radiation at relatively high frequencies. Assuming that all radiation is absorbed and converted to thermal energy, the spontaneous polarization of the sample changes and a pyroelectric current can be measured.^{1,5,6} This method is less sensitive to the effect of trapped charges. A further disadvantage is the uncertainty in the amount of energy actually absorbed which can lead to large errors. (3) The direct temperature ramp method, in which the poled sample is uniformly heated with a constant rate. The most important problem with this simple method is that thermally activated charges induced during the poling process are measured as an

indistinguishable part of the pyroelectric current.⁷ (4) The continuous temperature oscillation method that is specially designed for separating pyroelectric current from non-pyroelectric current is a modified version of the direct method.^{7–10} This method is based on the difference in the thermal relaxation time of polarization changes of pyroelectric current and that of the thermally stimulated current. In fact, the pyroelectric current is directly proportional to the derivative of the temperature with respect to time, while non-pyroelectric current is either constant or proportional to the temperature. When the sample is heated with a small sinusoidal temperature wave, an alternating current (ac) current is produced by the sample which amplitude and phase defines the ratio of pyroelectric and non-pyroelectric currents. The ac component in phase with the temperature wave is the non-pyroelectric current. However, if the ac component precedes the temperature wave by 90°, the origin is a pyroelectric current.^{7,8,10} The amplitudes of the non-pyroelectric (I_n) and pyroelectric currents (I_p) and the phase angle are given by

$$I_{ac} = I_{peak} \sin(\omega t + \theta), \quad (1)$$

$$I_{peak} = (I_n^2 + I_p^2)^{1/2}, \quad (2)$$

$$I_n = RT, \quad (3)$$

$$I_p = pAT\omega, \quad (4)$$

$$\theta = \tan^{-1}\left(\frac{I_p}{I_n}\right) = \tan^{-1}\left(\frac{pA\omega}{R}\right), \quad (5)$$

while R is a temperature coefficient of the non-pyroelectric current, A is the electrode area of the pyroelectric material, p is the component of the pyroelectric coefficient normal to the electrodes, ω is the angular frequency of the sinusoidal temperature component, and T its amplitude. This equation holds provided that measurements are carried out under constant stress and electric field so as to avoid piezoelectric, ferroelastic, and ferroelectric contributions.^{11,12} The frequencies involved in

this technique typically vary in the range of 0.2–0.02 Hz.³ However, some applications for pyroelectric materials may exist, in both sensing and energy harvesting domains, that require the characteristics of the materials over larger bandwidths, specially at lower frequencies. For instance, the human motion detectors are sensors of slow thermal processes such as approach or small motion of the human body, whose frequency spectrum is located in the region of the infra-low frequencies of 0.1–1 Hz.^{13,14}

In this paper, the experimental setup of a simplified version of the temperature modulation method is presented that is designed to measure the pyroelectric properties of the materials in the ultra low frequency spectrum, ranging 0.001–0.250 Hz. A novel signal processing route and phase angle analysis to characterize the pyroelectric coefficient of materials are presented. Detailed analysis of the test unit, sensitivity, accuracy as well as noise is discussed. The method is validated using LiTaO_3 single crystals and shown to perform very well at low frequencies.

II. TEST SETUP

A. Basic concept

A schematic diagram of the test setup is shown in Fig. 1. The measurement equipment consists of an aluminum Faraday cage equipped with a heat sink on top of which the test cell is located. A MCPE1-07106NC-S Peltier cooler receives sinusoidal signals from a power supply and generates the temperature modulation. A 1 mm thick copper plate is mounted

on top of the Peltier cooler for temperature uniformity. A thin layer of thermal conductive paste is used on the interface between the Peltier and the copper plate to ensure proper thermal conductivity. The pyroelectric sample is placed on the copper plate and probed with a gold plated pin electrode. The temperature sensor, a fast response (0.6 s, price less than 50 cents) MCP9701A SOT23 thermistor device from Microchip is mounted very close to the sample. The temperature control unit is based on the ATMEGA5 Xplained development board from Atmel. The 12 bit Xmegs on board ADC are used with 128× oversampling to create a 16 bit (± 15 bit) analog-to-digital converter (ADC) and still capable of running at an effective sample rate up to 1 kHz. Using this mode of the ADC together with an accurate 2.5 V (LM4040 device) reference voltage results in an overall resolution of $67.3 \mu\text{V/bit}$. The combination with the MCP9701 sensor device having a sensitivity of 19.5 mV/K makes it possible to control the temperature with 3.913 mK precision. The software is built such that this precision is always maintained, resulting in the smallest digital temperature disturbance possible. The 1 K temperature sine amplitude is done in 256 steps of 3.913 mK. The sine period is built with 1606 ($2\pi * 1 \text{ K}/3.913 \text{ mK}$) time steps, independent of the desired sine frequency (range 1 mHz–250 mHz). A temperature set point generator is realized that always uses the smallest temperature step possible. The same method is used to generate temperature ramping. The overall sine frequency or temperature ramping rate is controlled with the frequency of the (minimum temperature resolution) steps. The parameters for the temperature profiles can be programmed by Universal Serial Bus (USB) as virtual communication port.

FIG. 1. Schematic diagram of the set-up for the pyroelectric current measurement.

B. Temperature controller and noise analysis

A timer controlled ISR routine running at a fixed 500 Hz starts the ADC temperature measurement. The ADC ready flag starts updating the proportional-integral-derivative (PID) controller with the generated temperature profile as input. The 500 Hz frequency is based on half the maximum rate of the ADC and considered high enough for the sampling rate of the temperature control loop to cope with the Peltier thermal time constant. The result of the PID controller is checked for sign to operate the polarity switch for the Peltier. This polarity switch is simply built using 4 Avagos optically controlled SSR switches in bridge between the power supply output and the Peltier element. The modulus of the PID result is converted with the internal 12 bit DAC to a voltage that drives the Delta ES150-10 power supply as power amplifier for proportional control of the Peltier current. This combination is able to drive the Peltier with 40 W of bipolar power at 500 Hz sample rate.

The PID controller with 500 Hz sampling rate allows for a high closed loop bandwidth. The bandwidth can be controlled with the PID parameters to reduce closed loop temperature noise. Noise on the temperature translates to current noise from the sample. For frequency and phase measurement, this noise is easily canceled by the Fast Fourier transformation (FFT) convolution (shown in Sec. II D). For direct measurement of the temperature ramping current this could result in a too low overall signal to noise figures at low ramping rate. Therefore, a different set of PID parameters for sine modulation and temperature ramping is preferred. The temperature signal to noise ratio at the lowest 1 K amplitude, 1 mHz sine modulation is 30 dB or 0.03 K rms as measured in the 2 Hz sampled temperature data. The spectral noise is slightly folded, also due to aliasing, around the bandwidth (1 Hz) of Peltier controller. The pyroelectric current signal to noise ratio (SNR) is much lower due to its principle dT/dt character which give a 6 dB/octave rise of the noise with frequency.

The absolute current accuracy as specified for the Keithley electrometer in the current range is 0.2%. This accuracy affects the accuracy of P (Ipyro) but does not have an effect on the phase accuracy. The character of the noise on both temperature and current is mainly correlated due to the common source and the SNR on the current, being mainly a transformation of the noise on temperature, barely depends on the current amplitude. Basically, the convolution method (cross correlation) in the frequency domain multiplies the temperature-frequency spectrum and the current-frequency spectrum element by element resulting in a product of both individual SNRs. The SNR of the result is at least equal to the individual sum of both SNRs in dB. In fact, it will be higher because uncorrelated noise in the individual spectra is greatly reduced. For the current, the signal to noise ratio evaluates to 3 dB at the lowest frequency of 1 mHz. The applied correlation makes use of all recorded samples and therefore basically improving the overall SNR with the square root out of the total number of samples or $\sqrt{2000}$ per each fully logged modulation cycle at 1 mHz. This will give an overall SNR on the correlated frequency of 30 dB(temperature SNR) + 3 dB(current SNR) + 20 log($\sqrt{2000}$) resulting in at least 66 dB(1995 : 1). By

logging more than one full modulation cycle only (collecting more data-points) the overall SNR can be further improved.

C. Temperature and current sampling

The temperature controller determines the overall sampling rate when used in auto transmit temperature mode over the USB virtual port. Although the controller is capable of 500 measurements per second, the Keithley electrometer can only output 2 measurements per second when using the standard RS232 output as in the current study. Therefore, all measurements were performed using this sample rate, allowing frequency measurements up to $F_{sample}/2$ (Shannon) or 1 Hz. The temperature controller is used with the temperature measurement in average mode set to 250 for a 500 Hz/250 or 2 Hz effective sampling rate. Overall control for settings and file storage is done by *LabVIEW* software running on a PC or laptop. This software controls the settings of the temperature unit, has a graphical viewer for the output data (temperature and current) and user controlled logging. Receiving data from the temperature controller triggers the Keithley electrometer for new measurement. This pair of correlated data, both temperature and current are stored into a data file. Logging is done by starting the instrument and letting it acquire at least 1 complete period of the modulation frequency data. In case of ramping, the logging is started just before the ramp and stopped after reaching the final temperature. At 1 mHz with 2 Hz sampling, this results in a ~20 min measurement with 2000 data points. For higher modulation frequencies, the measurement time is proportionally shorter.

D. Offline data processing

The goal is to measure the phase difference between the pyroelectric current and the modulated temperature as well as the pyroelectric current amplitude. The collected raw data are used in the signal processing phase. This gives the researcher maximum possibility to process the data for new findings in multiple perspectives. Currently the raw data are imported in Mathcad but can be imported in any other mathematical program as well. The measured temperature and current are read-in as a vectored data set. From the acquired data, the maximum number of full modulation cycles is extracted to avoid errors due to incomplete cycles. This could otherwise result in foreign frequencies of phase-like modulation in the frequency domain. Both sampling rate and modulation frequencies are synchronized and known and full measurement cycles can be isolated exactly. The time related signals temperature and current are transformed to the frequency domain by using a FFT. By applying the complex conjugate on the current signal and performing convolution within the frequency domain, the correlation between the signals is calculated. The DC-component is removed from the result as it adds to the real component of the correlation. The phase between both signals is simply calculated by the arc tangents of the DC-free results.

With a signal to noise ratio of 30 dB on the temperature measurement and only 3 dB on the current measurement (see

PID controller) the accuracy of the extracting method results in a phase error of less than 0.25° at the low frequency of 1 mHz, proportionally improving with increasing frequencies (0.02° at 10 mHz). This method clearly eliminates the need for costly lock-in amplifiers for this application. The simulations show that even with negative signal to noise ratio on current the phase result is still less than 0.5° .

E. Phase correction for systematic errors

To calculate the actual phase angle between the pyroelectric current and the temperature signals, the FFT driven phase is corrected for heat-transfer and instrument delays.

The phase delay due to the heat transfer is first discussed. The overall control loop is given in Fig. 2.

The feedback element of the Peltier surface temperature is the MCP9701 with a thermal time constant of only 0.6 s. On top of the Peltier, a thin (1 mm) copper plate is used to create a uniform temperature signal as well as a smooth surface to mount the sample on. Copper with low thermal resistance and relative low thermal capacity acts as a nice material to be used as an additional filter for the noise on temperature, increasing the overall signal to noise ratio. This copper filter is kept outside the electrical PID loop as it otherwise gives rise to instability and canceling its effect as being part of the inside loop. The loop-gain is high enough (see Fig. 2) to reduce the Laplacian overall transfer function to

$$H_{transfer} = \frac{s \cdot \tau_{Plate} + 1}{s \cdot \tau_{Sensor} + 1}, \quad (6)$$

$$s = 2\pi i f. \quad (7)$$

The heat transfer function, $H_{transfer}$ is calculated based on the thermal time constant of the temperature sensor (τ_{Sensor}) and that of the copper plate τ_{Plate} . The thermal time constant of the plate is determined by comparing the recorded

temperature signal measured by the thermistor in the unit and the temperature of the plate monitored using an IR camera at different test frequencies. The final heat transfer phase delay, ϕ_{Heat} , is then calculated as

$$\phi_{Heat} = \arctan\left(\frac{\text{Im}(H_{transfer})}{\text{Re}(H_{transfer})}\right), \quad (8)$$

where $\text{Re}(H_{transfer})$ and $\text{Im}(H_{transfer})$ are real and imaginary components of the heat transfer function.

Finally, a small compensation is added for the overall instrument delay. As the overall sampling rate is kept constant at 2 Hz, the averaging of temperature inside the control unit and fixed sample delay of the Keithley electrometer adds a small compensation for this character. This contribution is characterized and compensated for in the following manner. The instrument delay, ϕ_{Inst} , consists of two components, delay of the electrometer and delay of the temperature control unit as follows:

$$\phi_M = t_M \cdot 2\pi f, \quad (9)$$

$$\phi_T = t_T \cdot 2\pi f, \quad (10)$$

$$\phi_{Inst} = \phi_M - \phi_T, \quad (11)$$

where ϕ_M and ϕ_T are the phase angles corresponding to sampling delays of the electrometer and temperature control unit, t_M and t_T are the sampling times of the electrometer and temperature control unit, f is the frequency of the thermal sine wave.

Therefore, the total phase delay, $\phi_{Correction}$, is calculated as the sum of the instrumental and heat transfer phase corrections, $\phi_{Correction}$, is added to the calculated phase shift based on the Fourier transformation,

$$\phi_{Correction} = \phi_{Inst} + \phi_{Heat}. \quad (12)$$

The actual phase is then calculated by adding the total phase delay, $\phi_{Correction}$, to the measured phase.

FIG. 2. Simplified temperature control loop.

TABLE I. Measured pyroelectric properties of $LiTaO_3$ chip at 25 °C and $\Delta T = 1$ °C.

Frequency (mHz)	Phase angle (deg)	I_{peak} (A)	$P_{measured}$ ($\mu C m^{-2} ^\circ C^{-1}$)	$P_{reported}$ ($\mu C m^{-2} ^\circ C^{-1}$)	Ref.
1	89.5	-2.75×10^{-11}	-175.1	Not reported	...
5	89	-1.35×10^{-10}	-171.8	175	⁸
10	88.7	-2.68×10^{-10}	-170.5	175	⁸
30	92	-8.3×10^{-10}	-176	Not reported	...
60	91	-1.65×10^{-9}	-175.1	172.7	¹⁶
100	97.9	-2.41×10^{-9}	-152	175	⁸
150	99	-3.90×10^{-9}	-163.2	Not reported	...
200	101	-5.64×10^{-9}	-176.3	160	⁸
250	104	-7.14×10^{-9}	-176.5	Not reported	...

III. MEASUREMENTS AND VALIDATION

The device and the data processing protocol were validated using $LiTaO_3$ single crystals whose properties are well documented and have low sample to sample variations. Fully poled commercial $LiTaO_3$ wafers of 5 mm $\times$ 5 mm $\times$ 0.2 mm were obtained from Infratec, Germany. The measured pyroelectric properties as a function of frequency from 1 mHz to 250 mHz at 25 °C are listed in Table I and compared to the reported values from the literature obtained by continuous temperature oscillation method.

The phase difference between the temperature and current signals is determined to establish which type of current is present. When both pyroelectric and non-pyroelectric currents are present the phase angle varies, and the fractions of current associated with pyro and nonpyro effects are calculated as

$$I_p = I_{peak} \sin(\theta) = pAT\omega, \quad (13)$$

$$I_n = I_{peak} \cos(\theta) = RT. \quad (14)$$

For the calculation of pyroelectric coefficient, the peak current (I_{peak}) is first measured and the pyroelectric coefficient is calculated as

$$p = \frac{I_p}{AT\omega} = \frac{I_{peak} \sin(\theta)}{AT\omega}. \quad (15)$$

Three samples are tested at each frequency. No dependencies of pyroelectric current to frequency are observed indicating a pure pyroelectric behavior for $LiTaO_3$. The measured phase shifts at all frequencies show values close to 90° indicating that the ac current has almost no non-pyroelectric component.⁸ The calculated pyroelectric coefficients in the range of 1–250 mHz are in agreement with the reported values from the literature (170 – $190 \times 10^{-10} \mu C m^{-2} ^\circ C^{-1}$).^{8,15,16} Therefore, it can be stated the accuracy of the test unit is high over the whole frequency range of 1–250 mHz.

IV. CONCLUSIONS

It has been shown that one can accurately determine the pyroelectric coefficient of materials by applying a simple digital signal processing method on the discrete sampled data obtained in temperature oscillation technique. Using the proposed approach, one can mathematically perform the FFT analysis to combine residual noise filtration and phase shift analysis between temperature modulation and current. The

phase shift is accurately corrected based on the instrument and heat transfer delays of the test unit. The method is proved to be suitable for testing pyroelectric materials in the ultra low frequency spectrum of 1–250 mHz without using costly lock-in amplifiers.

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