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Automated Measuring System for Studying the Temperature Dependence of Dielectric Spectra of Ferroelectrics

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Abstract **An automated measuring system developed by authors for studying the temperature dependence of dielectric spectra of ferroelectrics in the frequency range 12Hz-100kHz, the temperature range 80-450K and the amplitude of the measuring signal 5mV-1.275V has been presented. It is based on the GW Instek LCR-819 LCR Meter, the Measurement Computing USB-TEMP-AI temperature meter and an OWON ODP3033 programmable power supply. The temperature measurement resolution is 0.001K and the controlled rate of temperature change is from 0.001K/min to 1K/min. The control software, created in the LabVIEW graphical environment, makes it possible to carry out measurements in an automatic mode, present the results obtained in the form of graphs (Cole-Cole plot and frequency dependency of ε' and ε") and save the data as a file for further processing.**

Keywords—dielectric spectra, temperature dependence, ferroelectrics.

I. INTRODUCTION

The dielectric constant of materials ε, and especially its temperature ε(T) and frequency ε(f) dependence, is a very important parameter that allows one to reveal phase transitions, various relaxation mechanisms, etc [1]. Recently, many manufacturers have offered LCR meters that cover the frequency range from millihertz units to tens of gigahertz. However, not all of them are suitable for the study of ferroelectrics, and especially ferroelectrics-semiconductors, which is associated with large values of the dielectric loss, reaching hundreds. Respectively, bridge circuits for such measurements are not suitable. Therefore, to study the temperature dependence of dielectric spectra, it is necessary to select an LCR meter with the required parameters, and also to provide the measuring system with the ability to change the temperature within the required limits. Of course, it is possible to purchase a ready-made measuring system, for example [2], but such installations cost hundreds of thousands of dollars. Therefore, we have developed a measuring system based on an inexpensive LCR meter, a precision temperature meter module, and a programmable laboratory power supply. The software was created in the LabView graphical environment.

II. FEATURES MEASUREMENT OF FERROELECTRICS

Most of the methods currently used to study the dielectric properties of ferroelectrics are borrowed from the field of measurement of linear dielectrics. The specificity of these materials has led both to the improvement of known and to the development of fundamentally new methods [3]. However, the solution to this problem is still far from complete. The latter is explained by the fact that the process of polarization of the ferroelectric in the external field is very complex and depends on many factors, including the mode in which the measurement is conducted. As a result, even in the elementary analysis of the dielectric properties of ferroelectrics, and especially ferroelectrics-semiconductors, it should be borne in mind that the magnitude of polarization, and hence complex dielectric constant ε*, significantly, and not linearly, depends on the electric field strength (E), it can change dramatically depending on the temperature (T), and is usually a strong function of time (t) in a very wide range. In addition, for these materials, a significant role is played by the history of the sample (the degree of imbalance of the domain structure, which depends on the rate of temperature change; conditions of "annealing" before measurements; exposure time of the sample in a temperature range; stages of research, the impact of surface effects, etc.). Ferroelectrics-semiconductors of the $M_2P_2S_6$ family have the feature that the dielectric losses in them are quite high even far from the phase transitions, which is due in part to the significant through conductivity of the samples, which increases sharply with increasing temperature and photoactive lighting.

Therefore, to study materials of the $M_2P_2S_6$ family, the LCR meter GW Instek LCR-819 was chosen. It covers the low-frequency range, in which both types of effects are observed — those associated with the rearrangement of the domain structure and those caused by various mechanisms of low-frequency relaxation. The magnitude of the measuring field for this device is relatively small (from tens of millivolts), which excludes the contribution of strong nonlinearity of the samples, and also has (as our experiments showed) excellent stability and reliability over time.

III. EXPERIMENTAL SETUP

The block diagram of the automated measuring system developed by us is shown in Figure 1.

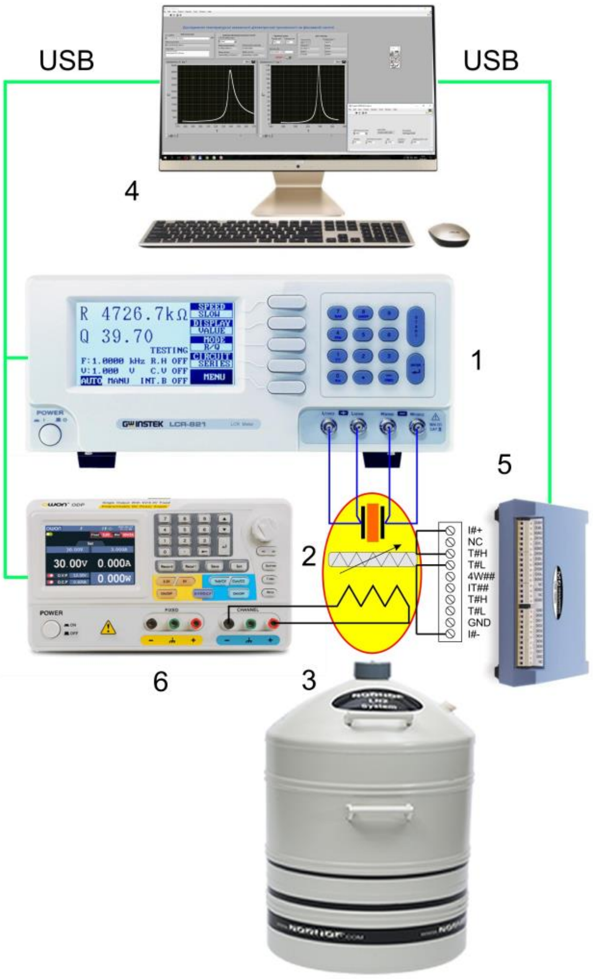


Figure 1. The block diagram of the automated measuring system for studying the temperature dependence of dielectric spectra of ferroelectrics

It consists of a GW Instek LCR-819 LCR Meter 1 (connected through RS-232↔USB converter), a cryostat 2 immersed in liquid nitrogen [4], a Dewar vessel 3, an IBM PC compatible control computer 4, a Measurement Computing USB-TEMP-AI temperature meter 5 (a high purity alumina ceramic case hand made platinum thermometer PT100/1509A of TDI Ltd. installed in a crystal holder is used as a temperature sensor with resistance tolerance at 0ºC 0.1% [5]), and OWON ODP3033 programmable power supply unit 6 to change the power of the heater.

The platinum thermistor is connected to the USB-TEMP-AI in a four-wire circuit to compensate for the resistance of these conductors. This is especially useful for the immersion-type cryostat, where the wires are 60-70 cm long. The sample is also connected to the LCR meter with four wires, which, in addition to the active resistance, makes it possible to compensate for the reactive components of the cable parameters, such as parasitic capacitance and inductance. For this, a special calibration mode for the LCR meter is provided, and the calibration data is stored in the device's memory.

The metrological characteristics of the measuring system are due to the accuracy of the LCR meter and temperature meter. The main characteristics of the GW Instek LCR-819 LCR Meter.

- Capacitance range - 10^{-5}pF ÷ 99999μF
- Dielectric loss tangent - $10^{-4} \div 10^{4}$
- Measurement accuracy - 0.05%
- Frequency range - 12 Hz ÷ 100 kHz (503 values)
- Measurement signal value - 5 mV - 1.275 V (with a step of 5mV)
- Measurement speed - 68 ms (minimum)
- Equivalent circuit - parallel / serial
- Connection interface - RS-232

Measurement Computing USB-TEMP-AI Module Parameters [6]:

- Temperature resolution - 0.001 K
- RTD Measurement Absolute Accuracy: ±2.9ºC at -200ºC and 0.27ºC at 100ºC
- Number of channels - 4
- Resolution - 24-bit
- Isolation - 500 V
- Speed - 0.5 s/channel

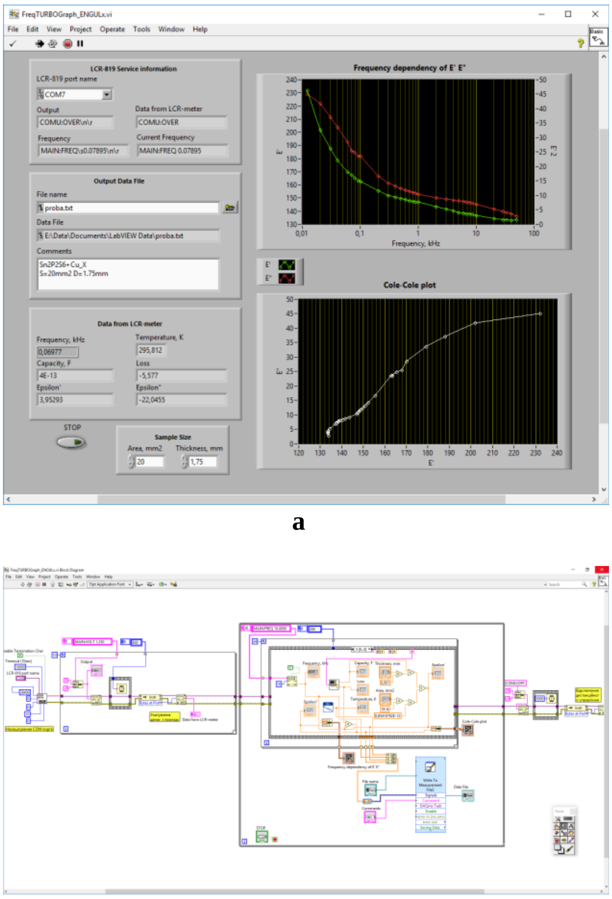
A linear change of the heater power (10W) using OWON ODP3033 programmable power supply allows us to change the heating and cooling rate in the range of 0.001 K / min to 1 K / min.

As it turned out, in the study of phenomena in the vicinity of phase transitions in ferroelectrics, especially for phase transitions of the first order, the monotonicity and rate of temperature change have the greatest importance. This is becose, at low cooling/heating rates, the smearing of the phase transition decreases (although it was expected that due to the phenomenon of overheating/overcooling, everything would be the other way around), and the violation of monotonicity can lead to an accidental transition to the paraelectric/ferroelectric phase. In this case, the absolute accuracy is not so important as the phase transition temperature is a function of the velocity. In addition, for most ferroelectric semiconductors, even a slight deviation from stoichiometry leads to a significant shift in the phase transition temperature.

As shown in our research, the ultra-low rate of temperature change makes it possible to discover brand new effects in the region of phase transitions in ferroelectrics semiconductors. Most likely, this is since at ultralow rates of temperature change, the system is closer to the state of thermodynamic equilibrium, which is also due to lower temperature gradients in the sample.

IV. CONTROL SOFTWARE

When creating automated measuring systems, the main problem is to create control software. Indeed, compared to connecting the system devices to a computer (which is not at all difficult due to the use of standard RS-232 or USB ports), the software component is almost always unique. If for most branded devices the use of SCPI (Standard Commands for Programmable Instruments, today part of the IVI Foundation [7]) is a standard, then cheaper analogs require individual programming, since they either do not support SCPI at all or partially.



LCR-819 port name: COM7

Output: COMU:OVER\n\r

Data from LCR-meter: COMU:OVER

Frequency: MAIN:FREQ\s0.07895\n\r

Current Frequency: MAIN:FREQ 0.07895

File name: proba.txt

Data File: E:\Data\Documents\LabVIEW Data\proba.txt

Comments: Sn2P2S6+Cu_X
S=20mm2 D=1.75mm

Frequency, kHz: 0,06977

Temperature, K: 295,812

Capacity, F: 4E-13

Loss: -5,577

Epsilon': 3,95293

Epsilon": -22,0455

Area, mm2: 20

Thickness, mm: 1,75

a

b

Figure 2. Front panel **a**, and the block diagram **b** of the automated measuring system for studying the temperature dependence of dielectric spectra of ferroelectrics.

The main programming feature of the LCR-819 meter was that the format of the output information on the RS-232 port did not always coincide with the technical description, which required the use of reverse engineering for the correct interpretation of the received data. The second feature was the conversion of the capacitance value from the mili-, micro-, nano-, pico-format to a numerical value. This is necessary for further processing of the obtained data. Using the formula of a flat capacitor, knowing the physical dimensions of the sample, the capacitance value is converted into the dielectric constant ε', and the tgδ into dielectric losses ε".

$$\varepsilon' = \frac{C \cdot d}{\varepsilon_0 \cdot S};\ \varepsilon" = \varepsilon' \cdot tg\delta, \tag{1}$$

where C – capacity, d – thickness, and S – area of the sample, $\varepsilon_0 = 8.8541878128 \cdot 10^{-12}$ F·m^{-1} the distributed capacitance of the vacuum.

The obtained ε 'and ε" values are displayed in the form of a graph of frequency dependence, as well as in the form of a Cole-Cole plot. For further processing, the data is saved to a file.

As we can see in Figure 2, we have chosen the LabView graphic system as the development environment. Its main advantage lies in the simplicity of connecting measuring devices, a large number of built-in functions for processing and visualizing the obtained data. In addition, it can be considered the de facto standard in the field of automation of a physical experiment.

The program only needs to specify the physical dimensions of the sample as input. To speed up the acquisition of the dielectric spectrum, the frequency is changed in a logarithmic scale (53 values in the range 12Hz-100kHz). Since at maximum accuracy the rate of capacitance measurement is on the order of one measurement per second, one spectrum is obtained in almost a minute. If necessary, the number of frequencies used can be either increased or decreased. The amplitude of the measuring field, as mentioned above, can be changed in steps of 5 mV. Our control program is freely available on GitHub [8].

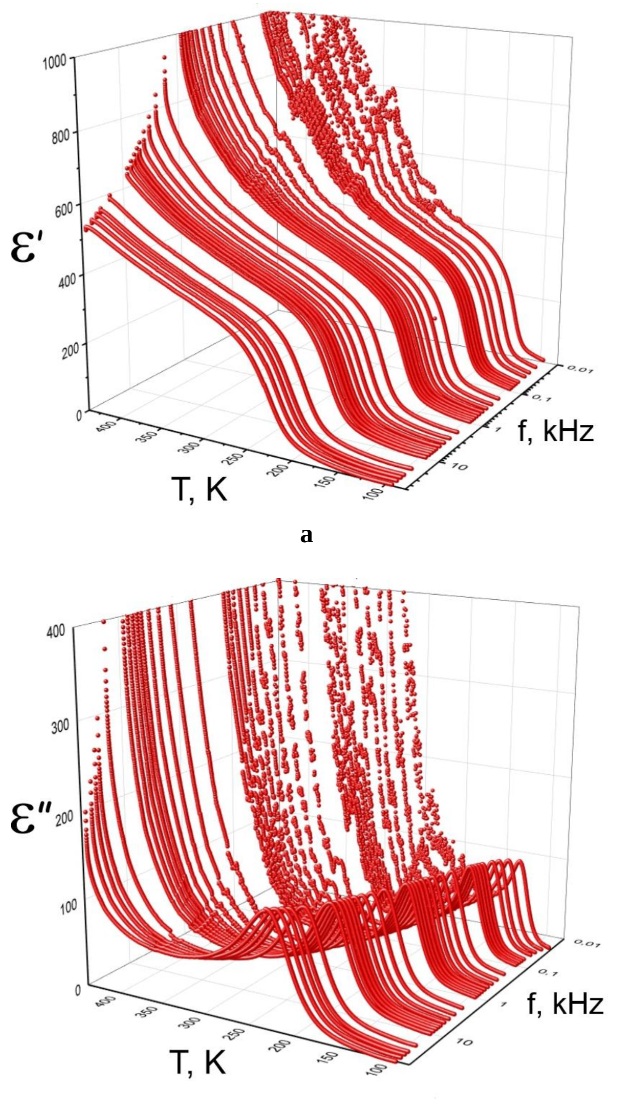
For linear heating and cooling of temperature, we also developed a control program for OWON ODP3033 programmable power supply that allows us to set the rate of temperature change in the range of 0.001K/min to 1K/min. As in the previous case, we had to use a non-standard procedure to initialize the remote control mode of the power supply using the external DLL library [9]. This program runs as a separate process, which allows temperature control independent of the dielectric measurement process.

V. TEST RESULTS

The automated measuring system has been created and used by us over the past few years to study materials of the $M_2P_2S_6$ family.

Using this technique, we confirmed the possibility of relaxation splitting of the second-order phase transition in $Sn_2P_2S_6$ crystals into a sequence of two second and first-order phase transitions, established the dependence of the smearing of the first-order low-temperature phase transition in $Sn_2P_2Se_6$ crystals with a change in the cooling rate, and many more nonequilibrium phenomena in crystals of the $M_2P_2S_6$ family.

To demonstrate the possibility of installation, Figure 3 shows the temperature dependence of real and imaginary parts of dielectric permittivity spectra of $Cu_{0.15}Fe_{1.7}PS_3$ mixed material.



a

b

Figure 3. Temperature dependence of real (**a**) and imaginary part (**b**) of dielectric permittivity spectra of $Cu_{0.15}Fe_{1.7}PS_3$ mixed material.

The anomalies observed on the graphs can be explained in the Maxwell-Wagner relaxation model, which is described by the formula

$$\varepsilon^* = \varepsilon_\infty + \frac{\Delta\varepsilon}{1+i\omega\tau} - i\frac{\sigma'}{\omega} \qquad (2)$$

where ε_∞ is the high-frequency permittivity, $\sigma' = \omega\varepsilon_0\varepsilon$ electric conductivity is calculated from the imaginary part of complex dielectric permittivity.

Such behavior is often found in heterogeneous systems in which components of the dielectric material have different conductivity [10].

VI. CONCLUSION

The automated measuring system for studying the dielectric spectra of ferroelectrics makes it possible to measure the electrophysical properties of ferroelectric semiconductors in a wide range of frequencies and temperatures. As shown in our research, the ultra-low rate of temperature change (0.005 K / min) allowed us to observe the effect of the nonequilibrium state of the system on the behavior of dielectric properties in the vicinity of phase transition. The immersion type nitrogen cryostat in combination with an automated measuring system gives a chance to carry out continuous measurements over several weeks, which in manual mode would be impossible. The use of inexpensive devices in the construction of the system made it possible to create an installation comparable to the characteristics of serial measuring systems, at a price one hundred times lower.

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