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AN AUTOMATIC SYSTEM FOR MEASURING THE FERROELECTRIC HYSTERESIS LOOPS USING THE MODIFIED SAWYER–TOWER CIRCUIT

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In the article, an equipment and practical application of an automatic system (created by the authors) for measuring the ferroelectric hysteresis loops using the Atmega328 microcontroller have been considered. The modern approaches to the classical Sawyer–Tower circuit's application was analyzed, and practical need for such development was proven. The schematic diagram and description of the main device's components were given. Test results on measuring the hysteresis loops in a barium titanate single crystal were presented, and they were compared with the data published earlier. Moreover, the results on measuring the ferroelectric hysteresis loops of an [110]-oriented $0.8Pb(Mb_{1/3}Nb_{2/3})O_3$–$0.2PbTiO_3$ single crystal in a temperature range of 120 – 300 K at frequencies from 2 to 50 Hz were presented.

Keywords: polarization, ferroelectric, hysteresis, Sawyer–Tower circuit

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УСТАНОВКА ДЛЯ ИЗМЕРЕНИЯ ПЕТЕЛЬ СЕГНЕТОЭЛЕКТРИЧЕСКОГО ГИСТЕРЕЗИСА НА ОСНОВЕ МОДИФИЦИРОВАННОГО МЕТОДА СОЙЕРА – ТАУЭРА

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В статье рассматривается устройство и практическое применение созданного авторами автоматического измерителя петель сегнетоэлектрического гистерезиса на основе микроконтроллера Atmega328. Проанализированы современные подходы к использованию классической схемы Сойера – Тауэра и показана практическая необходимость в выполненной разработке прибора. Приводится принципиальная схема и описание основных узлов созданного устройства. Представлены результаты тестовых измерений петель гистерезиса в монокристалле титаната бария, которые сравниваются с ранее опубликованными данными. Изложены также результаты измерения петель сегнетоэлектрического гистерезиса в монокристалле твердого раствора $(PbMg_{1/3}Nb_{2/3}O_3)_{0,8}$-$(PbTiO_3)_{0,2}$ (PMN-PT20) в температурном диапазоне 130 – 300 K.

Ключевые слова: поляризация, сегнетоэлектрик, гистерезис, схема Сойера – Тауэра

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Introduction

Polarization as function of the external electric field in ferroelectrics is a hysteresis loop; it is one of the main characteristics of ferroelectric materials. First studies on measuring hysteresis loops were carried out by Sawyer and Tower in the 1930s. The measurement method named after the researchers has been used ever since. The initial version of the circuit was a high-voltage generator series-connected to a flat sample capacitor and a reference capacitor connected to the deflection plates of the cathode ray tube. This measurement circuit was modified many times: with new methods proposed for compensating for parasitic effects in the sample [1], for measuring the output signal [2], and for special types of applied measuring voltage [3–5].

Sinusoidal or triangular measuring signal is used in the classical circuit. In this case, one of the main problems in measuring hysteresis loops is phase rotation of the measured signal due to parasitic effects in the sample, for example, high conductivity. This is expressed as distortion of the loop or as a loop appearing in the material where it should not, which is described in detail in [6]. This problem can be solved by using a measuring signal with a complex shape, for example, by applying pairs of pulses of each polarity. This method is called Positive Up - Negative Down (PUND) or sometimes the Double-wave method (DWM) [3] in literature. Polarization is switched during primary pulses (half-waves), and all effects that were not preserved when the external electric field was removed are measured during the secondary half-waves. There are more complex measuring signals but describing them is beyond the scope of our study.

Although the original measurement circuit is fairly popular due to its simplicity, it has many drawbacks. The most important one is that data collection is complicated. The only way to record the measured loops before widespread use of digital oscilloscopes was to photograph the oscilloscope screen or transfer the image to translucent paper (film).

One of the main advantages of a digital oscilloscope is that it can record the numerical data of the measured signal. While all modern digital oscilloscopes can perform this function, not all models necessarily allow to record the numerical values of measurements taken in *XY* mode required for applying the Sawyer–Tower circuit. In most cases, only the values from both channels of the oscilloscope as functions of time can be recorded, which is inconvenient and requires constant switching between the two modes. Besides, this method becomes much more complicated if special forms of the measuring signal are used in case of modified circuits. For example, there are relatively long pauses between half-waves in the double-wave method, in order to discharge the reference capacitor, and the oscilloscope may not have sufficient memory to record such a long measurement process.

Another considerable drawback in using an oscilloscope is that an operator has to be personally present, since most devices are not equipped with an automated data storage system synchronized with some external process. Hundreds of hysteresis loops may be measured in real experiments over long periods of time, for example, during long passes over the temperature range. In this case, some universal digital system should be used for data collection. Such systems are very common nowadays.

This study describes a system for data collection that is easy to manufacture. The system is based on widely available, inexpensive components. An example of the system's practical application is study of polarization of a PMN-20PT ferroelectric single crystal.

Circuit diagram and operating principles of the device

The proposed system for measuring ferroelectric hysteresis loops is based on the Atmega328 microcontroller, which is part of the Arduino Uno debug board. The circuit diagram of the device is shown in Fig. 1. The hysteresis loop can be measured with at least one output and one input analog voltage channel. The output channel in the diagram is based on the DAC8512 DAC chip, which has an integrated reference voltage source, a 12-bit resolution and an output voltage in the range from 0 to 4.095 V. An operational amplifier (op-amp) is used to expand the range of output voltages to ±10 V; it is a four-channel LM324 op-amp in this device. The supply voltage of the op-amp, which is 12± V, provides the necessary output signal amplitude. An AD1580 shunt diode with a stabilized voltage of 1.225 V, a stabilized current from 50 μA to 10 mA and an output impedance of 0.5 Ω was chosen as a reference source for shifting the output signal during range scaling. Trimmer potentiometers for range limits (R7) and zero (R5) have 25 revolutions, so output voltage scaling can be

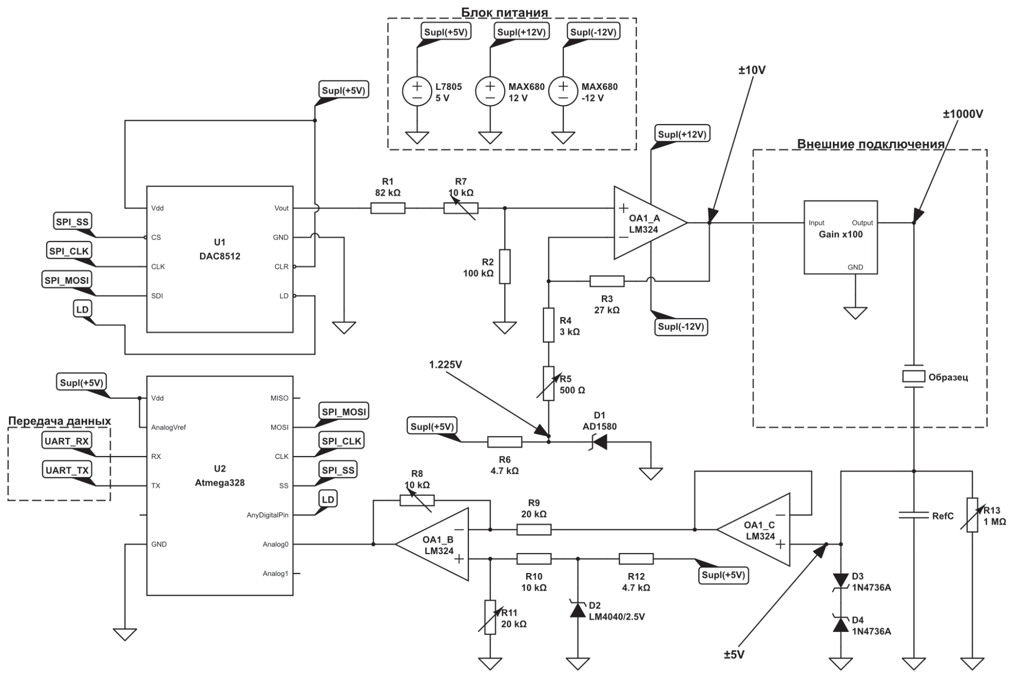

Fig. 1. Circuit diagram of main nodes of system for measuring hysteresis loops

adjusted with an accuracy no worse than 0.5%. The input channel measures the voltage drop across the reference capacitor and has a range of ±5 V. The built-in Atmega328 converter with 6 channels, a 10-bit resolution and reference voltage supplied to the AnalogVref input (5 V) is used as an ADC. Since the voltages in the test sample can significantly exceed the admissible input values, safeguards against sample breakdown (Zener diodes, TVS diodes, etc.) are installed on the input channel. Unfortunately, this solution has its drawbacks, for example, leakage currents of Zener diodes parallel-connected to the reference capacitor affect measurements like a phase-shifting variable resistor R13, which is not always acceptable. Better protection can be achieved by installing a high-linearity optocoupler, for example, HCNR201. The power supply unit is based on L78L05, L78L06 and MAX680 microchips and provides power supply of +5 and 12± V.

The diagram shows an example of external connections: a voltage amplifier with a gain of 100 and the installed sample (a flat capacitor with the given material). If the voltage amplifier is non-linear or has unstable parameters, its output voltage should also be measured. Another analog input channel (similar to the one described above) is constructed for this purpose, connected via a divider to the output of the high-voltage amplifier. In this case, the shape of voltage pulses from the output channel is selected based on the known behavior of the amplifier, and the voltages on the sample and on the reference capacitor are measured simultaneously by two input channels during operation.

The capacity for the reference capacitor is chosen so that the voltage drop across it remains within the acceptable range but at the same time occupies a significant part of the input voltage range. A block of capacitors is usually assembled to simplify the selection of this quantity; the capacitors can be connected to the circuit with either a manual wafer switch or with electromagnetic relays controlled by a microcontroller.

Resistor R13 is used to compensate for the phase shift between the applied voltage and the voltage generated on the reference capacitor if the sample has considerably high conductivity. The resistor can also be used as a current-measuring resistance. In the latter case, the reference capacitor is disconnected from the circuit, and the voltage drop across the resistor R13 is converted into the current flowing through it, so that the current hysteresis loops can be measured. Values of this resistance are typically tens of megaohms for compensated phase rotation and tens of hundreds of ohms for current measurements. It is convenient to use dual potentiometers as current-measuring resistors: one half of the device can be connected to another ADC channel of the microcontroller as a voltage reference divider; the microcontroller can then measure the resistance of the current-measuring resistor at any time for instant conversion of voltage into current.

The microcontroller can communicate with a computer by the RS232 standard (the circuit should be equipped with a UART-RS232 level converter for this purpose) or in virtual COM port mode via the USB interface built into the debug board, with the data transfer rate of up to 1 Mbps. The microcontroller can be programmed in the standard Arduino IDE, which does not require a separate programmer, or in AVR Studio using the I2C/SPI programmer.

Experimental studies

The ferroelectric hysteresis loops were measured on two samples: single crystals of $BaTiO_3$ (BTO) and $0.8Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.2PbTiO_3$ (PM-20PT) ferroelectrics.

Single crystals were prepared as follows: first, large samples were cut into plates with a [110] plane orientation and thicknesses of 70 μm for PMN-20PT and 600 μm for BTO, then the surfaces of the plates were polished with a DiaPro Nap R diamond suspension to a roughness less than 1 μm. Chromium-gold conductive electrodes 84 nm thick (Cr 4 nm, Au 80 nm) were deposited on both sides of the plates; Moorfield Minilab 080, a vacuum system for deposition of thin films, was used for this purpose. A Struers Accutom 50 machine was used for cutting, and a SuperNova X-ray diffractometer for measuring crystallographic orientation.

A single crystal of barium titatnate ($BaTiO_3$), which is a well-studied material, was chosen as a sample for testing and tuning the device constructed. Fig. 2 shows the obtained hysteresis loops (dark squares) and the signal of the secondary half-waves (light circles). A significant difference between the signals of the first and second pass indicates that polarization is switched in the sample, and the effect persists even when the external field is removed. The positive and negative parts of the loop are shifted relative to each other because the reference capacitor was discharged to zero after each half-wave was applied. The results obtained correspond to the hysteresis loops measured in [7] on the same material with good accuracy (of the order of 10–15%).

Measurements of hysteresis loops in PMN-20PT from our study [8] are given below as an example application of the device constructed. Numerous studies of polarization switching in PMNPT solid solutions [9, 10] point to a linear temperature dependence of the coercive electric field. The temperature model of hysteresis in ferroelectrics was described in [11]: the temperature dependence of the coercive electric field was found to be nonlinear. We assume that the temperature range in these experimental studies was not wide enough to observe non-linearity.

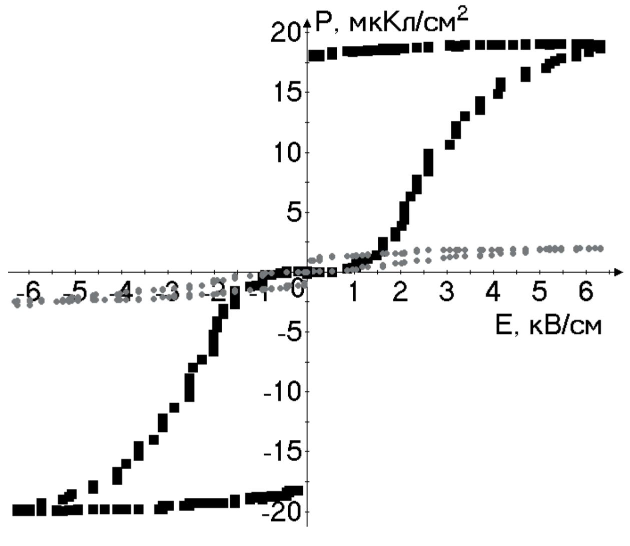

Fig. 2. Hysteresis loop obtained by double-wave method for $BaTiO_3$ single crystal; measuring system we constructed was used

The amplitude and frequency of the measuring signal were 400 V and 50 Hz, respectively; dark squares correspond to the signal of the primary half-waves, light circles to the signal of the secondary half waves

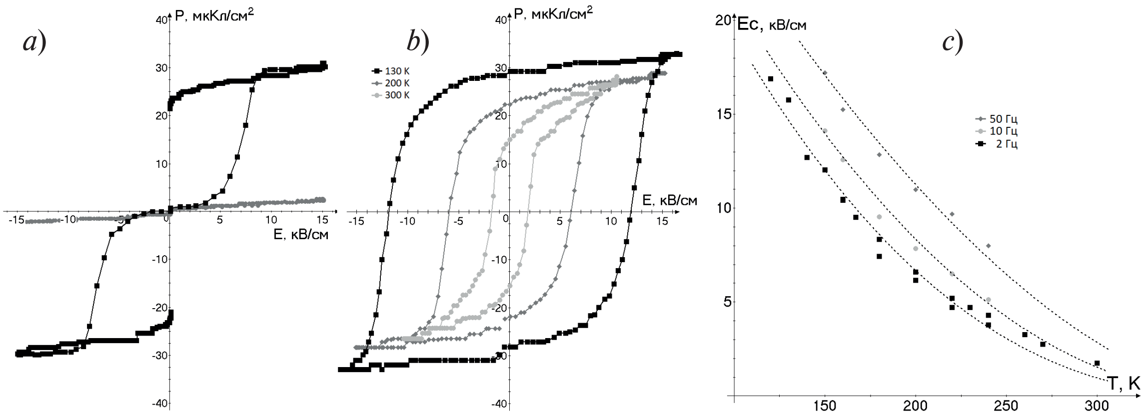

Fig. 3. Double-wave hysteresis loop in PMN-20PT single crystal (*a*) measured by DWM; hysteresis loops measured by conventional method at different temperatures (*b*). Dependence of coercive fields on temperature at different measuring frequencies (*c*). Dark dots in Fig. 3,*a* correspond to signals of the primary half-waves, light dots to signals of the secondary half-waves

Using the system constructed, we measured the ferroelectric hysteresis loops in the temperature range from 130 K to 300 K. Fig. 3,*a* shows quasistatic (measured at a frequency of 2 Hz) ferroelectric hysteresis loops at different temperatures. The double wave method (DWM) was used to increase the measurement accuracy. Fig. 3,*b* shows the results obtained for the primary (dark dots) and secondary (light dots) half-waves. A significant difference between the signals of the first and second pass means that the steps of the measured hysteresis loops are induced by polarization switching, while the parasitic effects are small and can be ignored. The magnitudes of the coercive electric fields were obtained using the measured hysteresis loops (Fig. 3,*c*).

The temperature dependences of the coercive electric fields we obtained are nonlinear, which corresponds to the model of ferroelectric hysteresis described in [11]. According to this model, the magnitude E_c of the coercive electric field is expressed as

$$E_c = E_h\left(1-\frac{T}{T_C}\right)^P, \qquad (1)$$

where E_h is the displacement field [12], T_c is the Curie temperature, p is the dimensionless constant.

Using expression (1), we performed regression analysis of the obtained dependences. The coefficients included in this expression were as follows (determined by the least squares method):

$$E_h = 40.1 \text{ kV/cm}; \ T_c = 380 \text{ K};$$

$$P = 2.4 \text{ at 2 Hz}; \ p = 2.1 \text{ at 10 Hz};$$

$$P = 1.7 \text{ at 50 Hz}.$$

It is difficult to estimate and compare the obtained coefficients, as our experimental study, carried out for a PMN-20PT single crystal using the above model, is the first in this direction.

Conclusion

We have developed and constructed a simple and effective device for measuring ferroelectric hysteresis loops, based on both the classical Sawyer–Tower circuit and the modified double wave method. We have given a circuit diagram of the main components of the measuring system. We have tested the system for a single crystal of barium titanate, yielding good accuracy of measurements. We have confirmed that the system we constructed could be used for a real scientific task, obtaining the hysteresis loops and values of coercive fields of a PMN-20PT single crystal in the temperature range from 130 to 300 K.

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