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Temperature dependence dielectric behavior of Ge-doped lead scandium tantalate single crystals

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Abstract

The doping of Pb_2SeTaO_6 with Ge ferroelectrics was produced by a high-temperature solution method. We measure dielectric constant, dielectric loss and conductivity in the temperature range −30°C to 200°C and frequency range 1 to 100 KHz. The value of dielectric constant of the Pb_2SeTaO_6 (PST) crystal remained the same after thermal annealing whereas they decreased after Ge doping in the phase transition temperature range of the PST single crystal. All samples were investigated for conductivity with increasing temperature.

Keywords: Dielectric constant, Ferroelectric, Paraelectric, Crystal lattice, Conductivity

Background

Complex lead-based oxides with the general formula Pb(B′, B″)O_3, where B′ = Sc^{3+} and B″ = Ta^{5+}, belong to the perovskite family and exhibit a ferroelectric-to-paraelectric phase transition with relaxor behavior. These materials show low-frequency dielectric dispersion, a high electrostriction coefficient and switchable pyroelectric property [1-4]. Pb_2SeTaO_6 crystals are of special interest for various practical applications in pyroelectrical detectors, electrochemical devices, capacitors, ultrasonic and medical devices and as materials for information data storage [5]. It has been shown that the state of ordering of the two B site cations in the perovskite structure can be modified by suitable thermal treatment (annealing in different atmospheres) [6] and doping with different elements [7]. In the present paper, the influence of the annealing treatment in air and doping with Ge on the dielectric constant, dielectric loss and ferroelectric conductivity of Pb_2SeTaO_6 (PST) single crystals will be discussed and analyzed.

Results and discussion

Figure 1 shows the temperature dependence of the dielectric constant of the non-annealed PST crystal at 10 kHz, 100 kHz and 1 MHz. For all frequencies investigated, at a temperature of about 15°C, we can see a clearly defined maximum in the dependence (T). The size of this diminishes with increasing frequency. It is evident that, at this temperature, there is a ferroelectric-to-paraelectric phase transition for non-annealed PST crystals. However, annealing and doping can shift the maximum of the temperature dependence in the dielectric constant, as can be seen from a comparison of Figures 2 and 3.

Annealing of the crystals (see Figure 2) leads to a shift of the phase transition to higher temperatures. In the temperature range of 0°C to 30°C, the values of the dielectric constant remain constant. After that, increases with increasing temperature up to 65°C for all measurement frequencies. Then, the dielectric constant decreases up to the highest temperature range examined.

Doping with Ge (see Figure 3) completely changes the dependence of (T) in comparison with non-annealed and annealed PST crystals. From −30°C to +30°C, the dielectric constant of the PST-Ge remains constant (approximately 1,000). At higher temperatures, it starts to increase. A strong increase can be seen at the lowest frequency (10 kHz). At the highest temperature examined, the values of the dielectric constant for PST-Ge crystals are 2,200, 1,500 and 1,100 at 10 kHz, 100 kHz and 1 MHz frequencies, respectively.

Sn^{2+} has the same electron configuration of the outer shell with that of Pb^{2+}. Thus, Sn^{2+} and Pb^{2+} are supposed to have a similar affinity to form lone pair electron and similar interactions with the nearest oxygen atoms. However, Sn^{2+} and Pb^{2+} have different radii. Therefore, doping with Ge should change substantially the tolerance factor of

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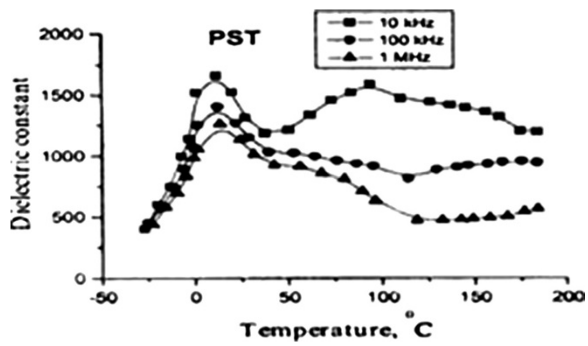

Figure 1 Temperature dependence of the dielectric constant of non-annealed PST crystals. At frequencies of 10 KHz, 100 kHz and 1 MHz.

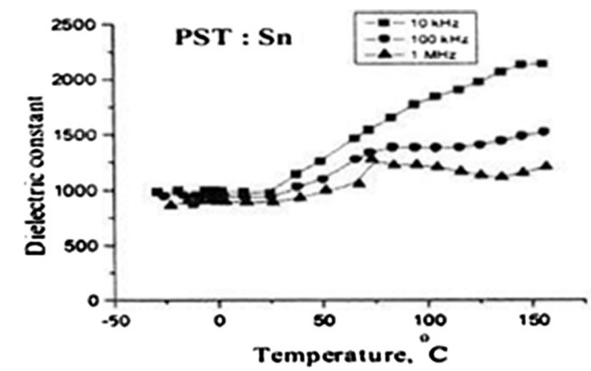

Figure 3 Temperature dependence of the dielectric constant of PST crystals doped with Ge. At frequencies of 10 KHz, 100 kHz and 1 MHz.

the perovskite, and the crystal lattice should be destroyed in the vicinity of the incorporated Ge atoms, thus affecting the ferroelectric properties.

Annealing of PST crystals and doping with Ge lead to a significant decrease in the values of the dielectric losses. At a 10-kHz measurement frequency, this is valid in the temperature range −30°C to +80°C; at 1-MHz frequency, it is valid from −75°C to the highest temperature examined. The values of the dielectric losses, tan(), for the annealed PST crystal and the PST-Ge crystals are approximately the same through the whole examined temperature range, while at 10 kHz frequency, they increase from 0.03 (at −30°C) to −0.14 (at 155°C).

Figures 4, 5 and 6 present the temperature dependences of the real part of the conductivity (Reσ) for the non-annealed, annealed and the doped PST-Ge single crystals, respectively. As shown, when the frequencies increase, the Reσ values become larger for all crystals. For non-annealed and annealed PST crystals, there is a maximum at the temperature of the phase transition for all frequencies. However, this maximum only very weakly present the annealed PST crystal. At a frequency of 10 kHz, the conductivity of the non-annealed PST crystals remains larger in comparison with the annealed and Ge-doped crystals over the temperature range of −30°C to +80°C. At higher frequencies, the Re values increase with increasing temperature for all samples.

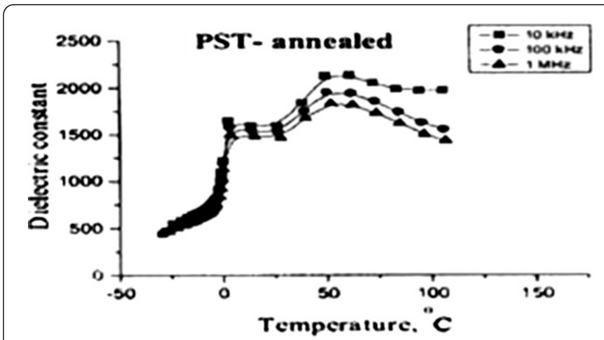

Figure 2 Temperature dependence of the dielectric constant of annealed PST crystals. At frequencies of 10 KHz, 100 kHz and 1 MHz.

Conclusions

Lead scandium titanate single crystals doped with Ge have been prepared using the high-temperature solution growth method by spontaneous crystallization. The values of the dielectric constant of PST crystal stay the same after thermal annealing whereas they decrease after Ge doping in the phase transition temperature range of the PST crystals. After that, the values of the dielectric constant of PST crystals become larger after thermal annealing as well as after doping with Ge. Furthermore,

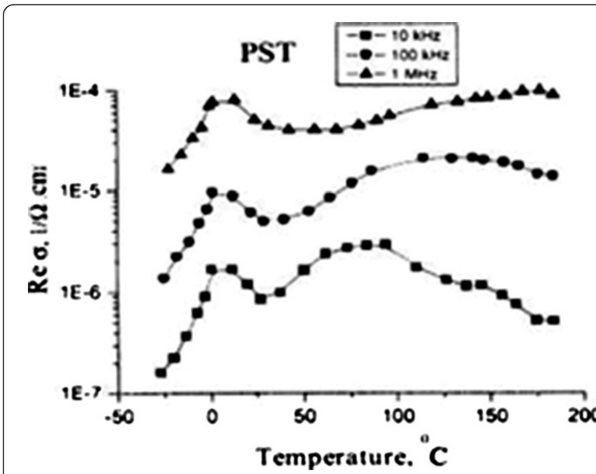

Figure 4 Temperature dependence of the real part of the conductivity of non-annealed PST crystals. At frequencies of 10 KHz, 100 kHz and 1 MHz.

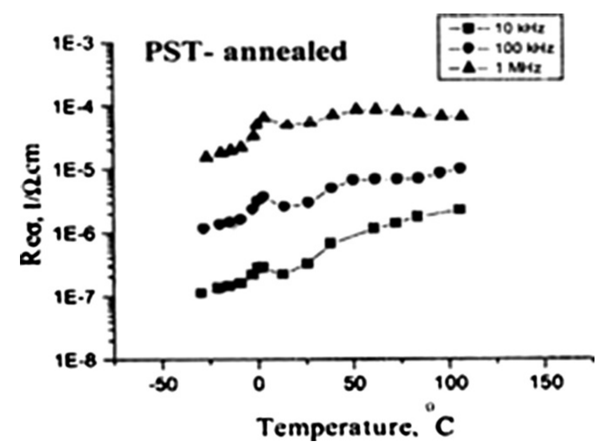

Figure 5 Temperature dependence of the real part of the conductivity of annealed PST crystals. At frequencies of 10 KHz, 100 kHz and 1 MHz.

the annealing and doping effects can shift the phase transition to higher temperatures. The conductivity, Reσ, increases with increasing temperature for all investigated samples.

The results presented on the influence of thermal annealing and Ge doping on the dielectric properties of PST ferroelectric crystals can be used to explain the relaxor behavior of such materials and for the optimization of their properties in the desired direction.

Methods

Pure crystalline perovskite Pb_2ScTa0_6 was synthesized by the solid-state reaction of stoichiometric amounts of PbO (99.999%), Sc_2O_3 (99.99%) and Ta_2O_5(99.99%) and further annealing for 48 h at 1,150°C in an oxygen atmosphere. Undoped and Ge-doped PST single crystals were grown by the high-temperature solution growth method using PbO/PbF_2-B_2O_3 flux (PbO/PbF_2-B_2O_3 = 0.75:0240.1). The flux was mixed with the PST powder and SnO_2 in a 10:1 ratio in the case of Ge-doped PST (the ratio was $PST/SnO_2 = 0.9:0.1$) and annealed at 1,230°C for 24 h in air. The temperature was then reduced to 950°C, and crystals with a typical size of $5 \times 5 \times 3$ mm were obtained. EDAX analysis established 3.4 vol.% of Ge in the PST crystals.

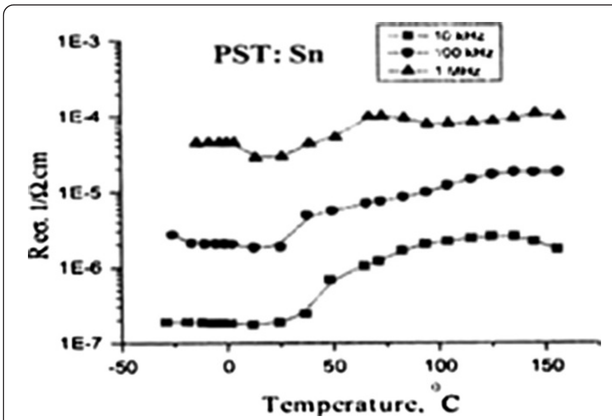

Figure 6 Temperature dependence of the real part of the conductivity of annealed PST-Ge crystals. At frequencies of 10 kHz, 100 kHz and 1 MHz.

For the dielectric measurements, the crystals were cut into flat-parallel plates, and silver electrodes were deposited on their opposite sides. The thicknesses of the samples were 1.100, 1.085 and 1.750 mm for the non-annealed PST, the annealed PST and the PST-Ge crystals, respectively. The dielectric measurements were made using Hewlett-Packard 4275A RLC bridges (Hewlett-Packard, Palo Alto, CA, USA) over a wide temperature range (−30°C to +200°C) and at 10 kHz, 100 kHz and 1 MHz.

Competing interests

The authors declare that they have no competing interests.

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