Soft Mode Behavior in Lead Germanate Studied by Terahertz Time-Domain Spectroscopy

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***Abstract*—We study temperature dependence of terahertz dielectric properties of ferroelectric lead germanate crystal. Observed soft mode behavior directly indicates transition from displacement to order-disorder type of ferroelectric in the vicinity of phase transition point.**

I. INTRODUCTION

Lead germanate $Pb_5Ge_3O_{11}$ (PGO) is a uniaxial ferroelectric with a transition temperature of T_c≈450 K at which the crystal changes from ferroelectric phase of C_3 symmetry to high-temperature paraelectric phase of C_{3h} symmetry. This structural phase transition of displacement type is associated with instability of the crystal against a particular normal mode of vibration A_{1g} known as a 'soft mode'. At the same time, experimental studies of PGO by Raman scattering and neutron scattering methods at temperatures 200 K÷500 K show presence of a central peak which is distinctive for phase transition of order-disorder type ferroelecrics [1]. Thus the mixture of two types of ferroelectic behaviour is observed in the vicinity of phase transition temperature.

Low-frequency terahertz time-domain spectroscopy is an effective means of polar lattice modes investigation. It allows studies of soft mode behaviour with high accuracy compared to Raman scattering in a wide range of temperatures.

We report on direct measurements of phase transition in lead germanate using terahertz spectroscopy techniques. We investigated *x*-cut samples of PGO crystals with a thickness of ~280 μm. Before the measurements the PGO samples were poled to a single domain by application of a z-directed electric field of ~16 kV/cm. Samples temperature varied from 297 to 443 K and was controlled with an accuracy of 0.5 K at reference points.

II. RESULTS

Imaginary part of dielectric susceptibility of lead germanate along *z* (polar) axis is shown on Fig. 1. The analysis of experimental data shows that phase transition has soft mode character, that is $\omega_0^2 \sim |T - T_c|$ for $|T - T_c| > 60\text{K}$, and $\omega_0^2/\gamma \sim |T - T_c|$ for $60\text{K} > |T - T_c| > 10\text{K}$ (ω_0 is the soft mode frequency, γ is the damping constant). The damping constant (full width at half maximum peak) increases rapidly and the soft mode becomes overdamped above ~380 K. At $|T - T_c| < 10\text{K}$ the soft mode becomes purely relaxational. The theoretical implications of our experimental observations agree well with the modern conceptions of structural phase transitions [2].

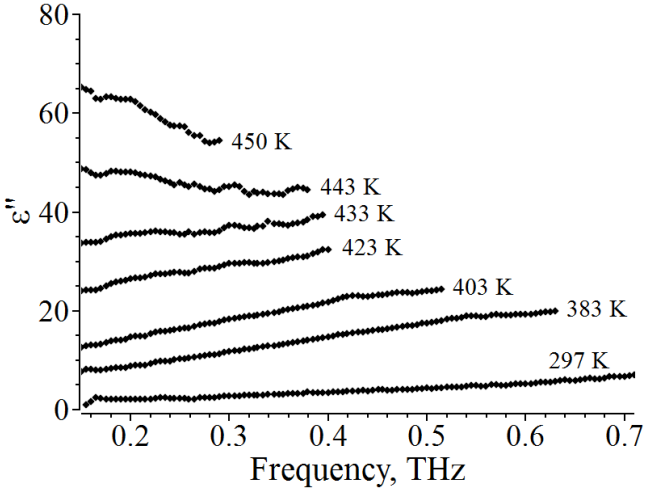

Fig. 1. Imaginary part of dielectric susceptibility of lead germanate in terahertz range at temperatures from 297 to 450 K. The absence of experimental data at higher temperatures is related to the excess of absorption over the dynamic range of the spectrometer.

Temperature dependence of ω_0 and γ shows that dielectric susceptiblity behavior is well explained with the model of coupled oscillators. It assumes linear coupling of the soft vibrational mode and independent relaxational mode. As we approach T_c (T>380 K) the vibrational mode softens and its low-frequency part overlaps with the relaxational mode. Interaction of these modes leads to creation of an overdamped hybrid mode which is mainly relaxational in the vicinity of T_c.

Thus for temperatures of $|T - T_c| > 60\text{K}$ PGO behaves as displacement type ferroelectric and for temperatures of $|T - T_c| < 60\text{K}$ as a typical order-disorder ferroelectric.

REFERENCES

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