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Nonlinear electromechanical behaviour of KDP near its phase transition

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Abstract

Potassium dihydrogen phosphate (KDP) undergoes a ferroelectric phase transition at 123 K. The linear elastic and electromechanical properties are well known. This paper deals with simultaneous measurements of the transit time and the attenuation variations of pulsed ultrasound in parallelepiped samples of KDP using a picosecond sampling oscilloscope. These variations are introduced by low frequency electric fields. The results can be used to determine complex nonlinear piezoelectric coefficients above and below the phase transition of KDP.

Keywords: Ferroelectrics; Acoustic properties; Elasticity; Phase transitions; Ultrasonics

1. Introduction

Several precision methods for the determination of the sound velocity of ultrasound pulse waves in solids are known. We wanted to have low jitter for the measurements with a digital sampling oscilloscope. Thus we developed a new measurement arrangement using a fast broadband oscilloscope HP54750, a burst generator Kontron 8550 and an amplifier chain with a MOSFET power amplifier followed by a tube amplifier. The amplifiers are of our own design. A detailed analysis of this measurement arrangement is given in Ref. [1].

The measurement equipment for the determination of nonlinear elastic and electromechanical coefficients [2,3] was improved in this way. We apply time dependent uniaxial stresses or electric fields to samples of solids that are examined by pulsed ultrasound. The nonlinear elastic coefficients can be obtained from the stress derivatives of the sound velocity whereas the electric field influence on the sound velocity delivers information about nonlinear electromechanical properties. We investigated potassium dihydrogen phosphate (KDP) near its ferroelectric phase transition temperature at 123 K. Petrakov et al. [4] determined nonlinear piezoelectric coefficients of KDP at room temperature. Measurements of the nonlinear piezoelectric coefficient d_{316} with a mechanical resonance method were carried out by Sysoev et al. [5].

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2. Experimental procedure

The KDP samples for the nonlinear electromechanical measurements had parallelepiped form. Axis cuts with 4 mm length in z-direction and 8 mm length in x- and y-directions were prepared. The samples were parallel and flat with an accuracy of 10 μm or better. The ultrasound was generated with y-cut quartz transducers of 20 MHz center frequency. The transducer for the KDP crystal was attached with silicon rubber. The measurement equipment for the determination of nonlinear elastic and electromechanical coefficients is described in detail in [6]. A low time jitter is especially important for the high resolution evaluation of small time shifts necessary for the determination of nonlinear effects. A programmable pulse generator followed by a two stage costum-built amplifier [1] produces 200 Watts pulse power at 20 MHz with a jitter of only 200 ps. The amplifiers are switched off when no pulses are generated. The signal-to-noise ratio during the reception of the echoes is improved in this way. The digital broadband oscilloscope HP 54750 has a very good time resolution of less than 8 ps. A computer is connected via IEEE to the oscilloscope for data recording.

3. Results and discussion

The following procedure was used for the measurements of nonlinear electromechanical effects. Approximately one cycle of the first echo r.f. pulse was measured using the sampling oscilloscope. The slowly varying external electric

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field of triangular time dependence induced delay time and amplitude variations. The r.f. cycle as part of the sampled signal is measured in dependence of this external electric field. The change of the sound velocity of a transverse wave along the z-axis polarized in [110]-direction due to an electric field in z-direction in KDP was treated in [7,8]. But the sound attenuation becomes field dependent below the phase transition. The true nonlinearity could not be measured with the method used in these former investigations.

We present in this paper measurements of sound velocity and attenuation changes due to an electric field in KDP with the above mentioned geometry near the phase transition of this crystal. All measurements were performed with symmetric triangular shaped electric fields. Some of the raw data taken with the new measuring system including the HP 54750 are depicted in Fig. 1. The signal sampled at 16 points of a 20 MHz echo cycle with fixed oscilloscope delay are presented in dependence of the triangular shaped external field. The resulting 200 sinusoidal curves per external field period are drawn in a three dimensional plot. The phase shift follows nearly linearly the triangular field in the top picture. The maximum field was 326 V/cm at a temperature 2 K below the phase transition temperature T_c in this case. The bottom part of Fig. 1 shows the relations 12.7 K below the transition temperature. Here the phase shift is no longer proportional to the triangular field. The raw data are processed in the following way. The r.f.-cycles are fitted using the program MATHEMATICA. The relative velocity change $\Delta v/v$ is calculated from the phase shifts. The calculated data are presented in Figs. 2–5. Fig. 2 is taken 0.3 K above T_c. It shows the sensitivity of the method, relative velocity changes smaller 10^{-4} can be clearly detected. A small field of 246 V/cm maximum field strength is enough to produce an effect. Fig. 3 shows the result 2 K below the transition and Fig. 4 that 3.7 K below T_c. The velocity follows in both cases still linearly the triangular field, but 3.7 K

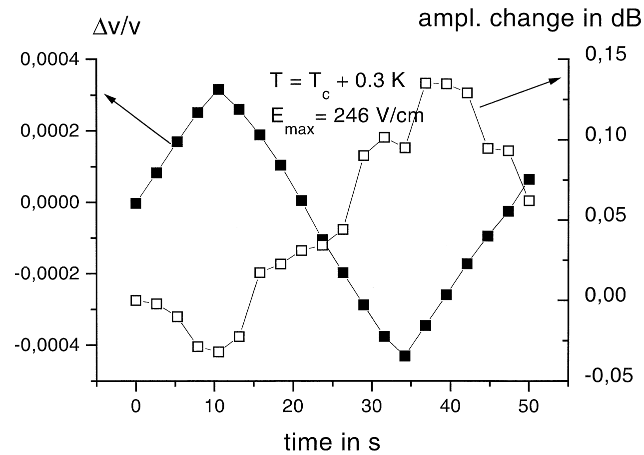

Fig. 2. Relative velocity and amplitude variations due to a triangular shaped electric field with the amplitude E_{max} applied in the z-direction. Sound wave along z, polarization along [110]. $T=0.3$ K above the phase transition temperature T_c.

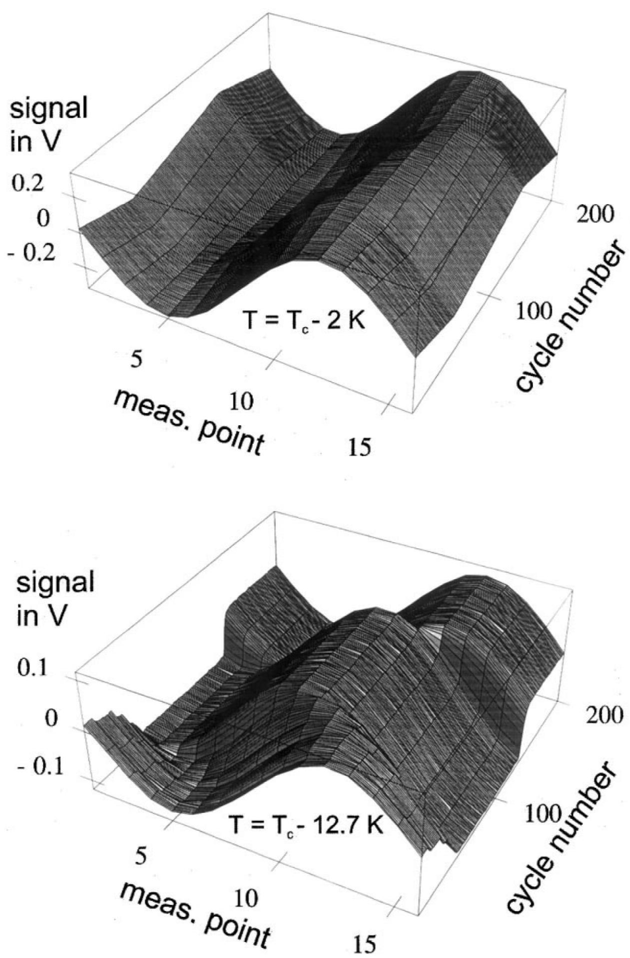

Fig. 1. Raw measurement data for a r.f. cycle part in the first pulse echo at different temperatures. Sound wave along z, polarization along [110]. The sample is exposed to a triangular shaped electrical field in the z-direction. Further explanation in the text.

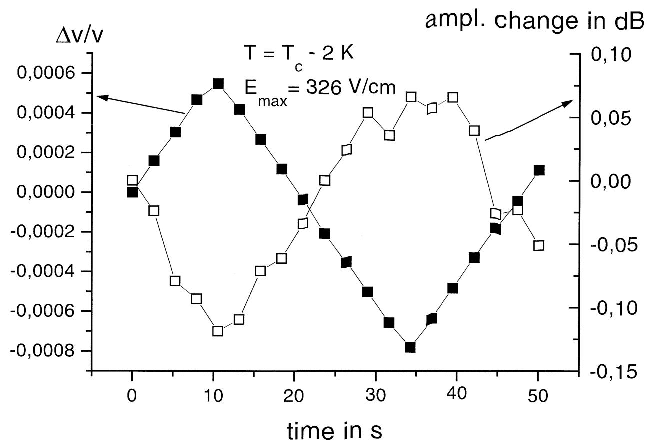

Fig. 3. Relative velocity and amplitude variations due to a triangular shaped electric field with the amplitude E_{max} applied in the z-direction. Sound wave along z, polarization along [110]. $T=2$ K below the phase transition temperature T_c.

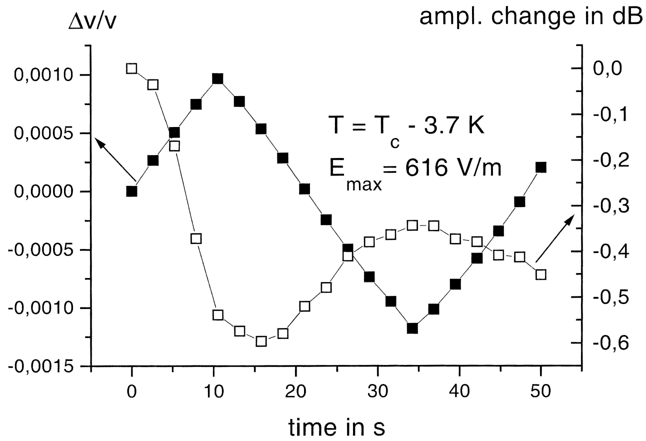

Fig. 4. Relative velocity and amplitude variations due to a triangular shaped electric field with the amplitude E_{max} applied in the *z*-direction. Sound wave along *z*, polarization along [110]. $T=3.7$ K below the phase transition temperature T_c.

below T_c the attenuation effect is not reversible. This is perhaps due to domain freezing [9]. The relation 12.7 K below the transition can be seen in Fig. 5. The steplike velocity change and the strong amplitude change are surely due to domain switching. An additional measurement of hysteresis loops gave a coercive field strength of about 700 V/cm at this temperature.

4. Conclusions

The measurements show the possibility to measure the influence of a slowly varying electric field on the velocity and attenuation of a transverse acoustic wave in *z*-direction with high precision. The improved measurement setup allows a good separation of velocity and attenuation effects. The change of the attenuation at temperatures below the transition temperature T_c can be perhaps attributed to the ferroelectric domain structure. The results above T_c show that nonlinear electromechanical effects have also a varying imaginary part expressed by the electric field dependent attenuation. Therefore an extension to the phenomenological theory of elastic and electromechanical nonlinearities [10] should be made. Far above the phase transition the following formula for the nonlinear piezoelectric coefficient e_{345} holds [7]:

$$e_{345} = d_{36}c_{456} - 2\varrho v^2[d_{36} - \Delta v/(v\ \Delta E)]. \quad (1)$$

In formula (1) d_{36} denotes the linear piezoelectric coefficient; ϱ the density; c_{456} the nonlinear elastic stiffness; and $\Delta v/(v\ \Delta E)$ the velocity variation due to the electric field variation ΔE. The nonlinear piezoelectric coefficient e_{345} was determined in Ref. [7]. The transducer arrangement and field direction is described in the Results and discussion section of the present article. Formula (1) is only valid for small attenuations. Therefore we mention this formula for completeness only. A better calculation and interpretation of the nonlinear piezoelectric coefficient would be possible if the mentioned extensions to the phenomenological theory of elastic and electromechanical nonlinearities would be done. The work on both the experimental and theoretical aspects of the determination of the nonlinear electromechanical and nonlinear elastic properties of KDP is continued.

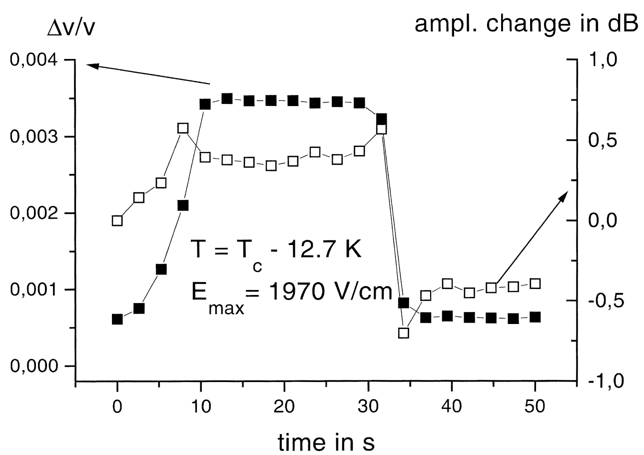

Fig. 5. Relative velocity and amplitude variations due to a triangular shaped electric field with the amplitude E_{max} applied in the *z*-direction. Sound wave along *z*, polarization along [110]. $T=12.7$ K below the phase transition temperature T_c.

Acknowledgements

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