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NONLINEAR ELASTIC PROPERTIES OF DOPED ALKALI HALIDES AND KDP[1]

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A new method for the determination of elastic nonlinear coefficients is described. Uniaxial mechanical stress is generated by means of a piezoelectric actuator. The uniaxial stress generates transit time variations of pulsed ultrasound in the sample. These variations are recorded with a computer controlled measuring system.
First measurements on Ca-doped NaCl shows a difference of nonlinear elastic coefficients for different Ca-concentrations, although the linear coefficients are the same. Nonlinear elastic properties of KDP in a rather wide temperature region are presented.

1. Introduction

The motivation to perform measurements of elastic nonlinearities in solids is caused by different reasons. The material behaviour at high mechanical stress is of particular interest for technical applications. The thermal expansion may be explained using nonlinear elastic coefficients. The influence of defects on nonlinear properties should be investigated. Phonon-phonon and phonon-photon interactions include nonlinear processes. The occurence of harmonics and parts of the attenuation of sound waves are due to nonlinear elastic effects. Some further fields are acoustic amplification and phonon echoes. Model calculations for simple crystal structures like in alkali halides are possible.

There are different methods for the determination of nonlinear elastic coefficients. The harmonics of the sound wave can be found using quartz transducers [1], capacitive sensors [2], or the diffraction on an "ultrasound lattice" generated with standing sound waves. These methods are not very precise because of the need of accurate amplitude measurements. Most of the coefficients are only available in combinations. The deviation from Hooke's law in the static experiment has the same disadvantage. Thus we favoured the measurement of the change of the ultrasound wave due to applied mechanical stresses and pressures.

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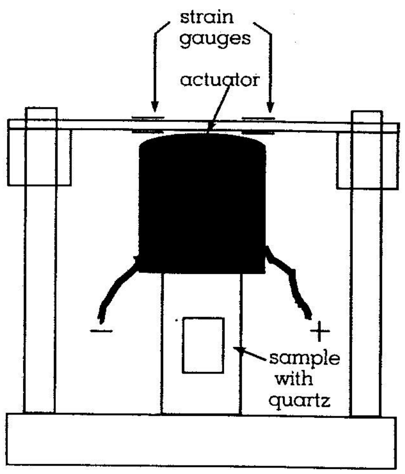

Fig. 1. Principle of the apparatus for the generation of uniaxial stresses

2. Experimental

The principle of the mechanical apparatus for the application of uniaxal stresses is shown in Fig. 1. It consists of two circular discs made from stainless steel. The top disk has a thick part at the radius and a thin inner part forming a membrane for the force measurement. Both disks are connected via three rods from steel or invar. The force is generated by an actuator that expands when a voltage is applied. The dilatation with free ends has an amount of 20 μm at 1 kV voltage. Thus the ultrasound sample with dimensions of about 1 cm is exposed to uniaxial stress. To achieve a nearly pure stress state a sample length of 10 mm and lateral dimensions of 5 mm are preferable.

This apparatus was mainly used for a broad temperature region down to liquid nitrogen. It was housed in a brass vessel with a heater and temperature control resistor and placed in liquid nitrogen dewar in the vapour phase. Different rod materials were tested to compensate the different temperature expansions of the central part consisting of sample and actuator and the three rods. Two invar rods and one of a definite stainless steel gave the best performance. The force measurement was done with four strain gauges arranged at the membrane of the top disk. The resulting strain measuring bridge was calibrated against a commercial load compression cell 13E500N0 from Sensotec with a maximum compression load of 500 N. This load cell worked very well in another more rigid apparatus for room temperatures too. The electronic setup is displayed in Fig. 2.

The ultrasonic pulse method used in this experiment is similar to the Papadakis method [3]. The TTL-pulse generator hp 3324A delivers a signal with a frequency that is approximately one hundred times higher than the inverse transit time between two ultrasound echos and is immediately divided by 100 to reduce phase noise. The master pulse is shaped following a divider 1/100 and has a variable length of 1 μs. A pulse selector consisting of a variable delay and a TTL gate produces very stable trigger pulses with a distinct delay from the master pulse. The master pulse was initially used to drive a high frequency generator with a frequency of 20 MHz. A better performance could be reached with a fast high voltage pulse amplifier and a pulse width of 50 ns. Filters were used suppress higher harmonics. The impuls echo train from the ultrasound sample with a sound path length of 5 mm is amplified with a band pass high frequency amplifier and reaches a digital sampling oscilloscope MFA 105. This instrument has a gate width of 150 ps and works for this method in a regime with a minimum fixed delay. A second slow channel of this oscilloscope watches the force signal from the strain gauge. Both outputs are taken directly from the 12 bit AD converters and send to the computer.

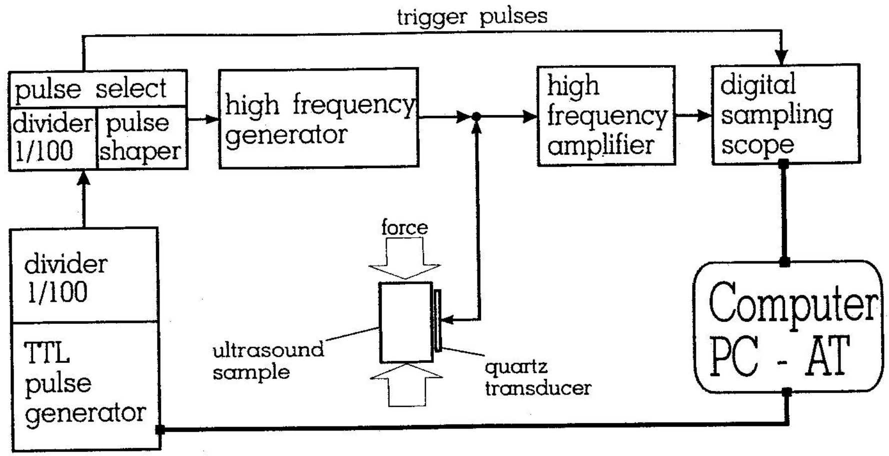

Fig. 2. Setup for the measurements of velocity variations due to external mechanical stresses

The trigger impuls is set to a zero crossing point of one hf echo. When the external force is applied the transit time variation causes a amplitude variation of the oscilloscope output. If the signal amplitude is not dependent from the mechanical stress the transit time variation can be recorded. This procedure was used in similar forms from several authors e.g. [4] with analogue sampling oscilloscopes. The resolution of the transit time variations is 10^{-6}to10^{-7}.

Automatic measurements in dependence of the temperature are possible too, because the TTL generator is computer controlled via IEEE. The frequency can be set permanently to a value that provides the zero crossing point. A similar method was proposed by Sarras and Kabelka [5].

We used an alternating rectangular voltage producing an uniaxial stress of the same shape. This allows the compensation of drift effects. The signal to noise ratio was enhanced by online summation of the captured data. The period of the applied mechanical stress was about 10 seconds and the sampling rate about 1 kHz. 250 data points were collected at each measurement run.

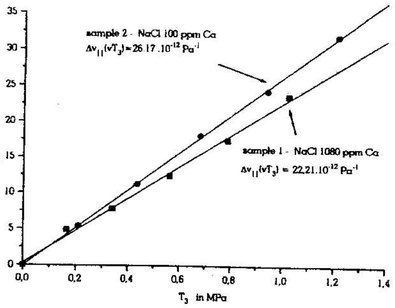

Fig. 3. Relative velocity variation $\Delta v_{-}/(v_0 \Delta T_3)$ related to the stress versus stress amplitude T_3 with stress parallel to the sound wave polarization

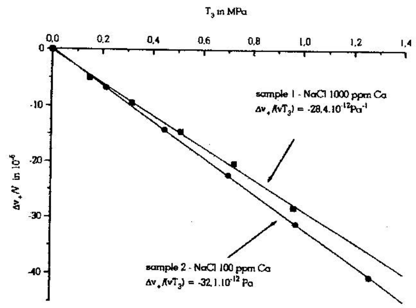

Fig. 4. Relative velocity variation $\Delta v_{+}/(v_0 \Delta T_3)$ related to the stress versus stress amplitude T_3 with stress perpendicular to the sound wave polarization

3. Results and discussion

First investigations were performed using samples of cubic NaCl doped with Ca at room temperature. We present results of measurements with transverse waves. The samples were cut in crystallographic axis directions. The sound direction was always perpendicular to the applied stress. The results for two samples with different Ca--content are displayed in Figs. 3 and 4.

Figure 3 shows the transit time variations when stress and sound wave polarization are parallel. The transit time variation increases linearly with the stress amplitude. The effect is larger for the sample with less calcium. A similar result was found for the case when the applied stress is perpendicular to the ultrasound polarization. Fig. 4 shows a negative slope of the transit variation versus stress amplitude. Sample 2 with 100 ppm calcium has again a larger absolute slope than sample 1 with 1000 ppm Ca. We took care to apply maximum stresses below the yield point. This yield point shows a concentration dependence [6]. The linear elestic value, the transverse velocity is not concentration dependent and is found to be $v_0 = 2407$ m/s. The exact nonlinear coefficients must be calculated in a procedure developed by Brugger [7]. The values for the density ϱ_0 and for the linear stiffness coefficients s_{11} and s_{12} are taken from [8].: $\varrho_0 = 2168$ kg/m^3, $s_{11} = 3,07 \times 10^{-11}$ Pa^{-1} and $s_{12} = -0.85 \times 10^{-11}$ Pa^{-1}.

We get two of the nonlinear elastic coefficients following Brugger:

$$c_{166} = \frac{2\varrho_0 v_0^2[s_{11}(B_{||} + s_{11}) - s_{12}(B_{+} + s_{12})]}{2s_{12}^2 - s_{11}(s_{12} + s_{11})} \tag{1}$$

$$c_{144} = \frac{2\varrho_0 v_0^2[2s_{12}(B_{||} + s_{11}) - (s_{12} + s_{11})(B_{+} + s_{12})]}{s_{11}(s_{12} + s_{11}) - 2s_{12}^2}. \tag{2}$$

The letter B denotes the relative velocity variation $\Delta v/(v_0 \Delta T_3)$ related to the stress T_3, the subscripts $||$ and $+$ indicate if stress and polarization are parallel or perpendicular. The result for c_{166} differs from $c_{166/1} = -61.27$ GPa for sample 1 with 1000 ppm Ca to $c_{166/2} = -65.39$ GPa for sample 2 with 100 ppm Ca. Note that the nonlinearity for the sample with low content of calcium is higher. Values from literature [8] for pure NaCl vary from -58 GPa to -61 GPa. The values for c_{144} obviously have an great error: $c_{144/1} = -3.74$ GPa for sample 1 with 1000 ppm Ca and $c_{144/2} = -3.00$ GPa for sample 2 with 100 ppm Ca. The coefficients in [8] range from 7 GPa to 29 GPa. The deviation for $c_{144/2}$ is partially due subtracting two nearly equal values with errors in the numerator in equation 2.

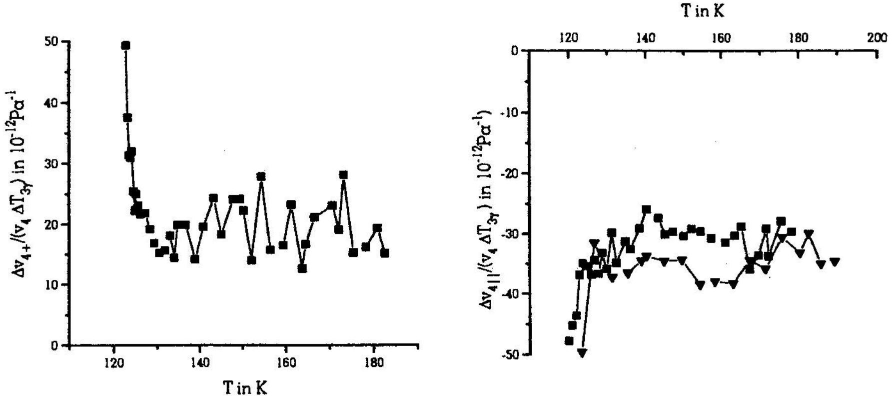

Fig. 5. Temperature dependences of the relative velocity variation of transverse ultrasound along the z-axis in KDP due to uniaxial stress in the [110]-direction, on the left hand side with parallel stress and polarization, on the right hand side with perpendicular stress and polarization [9]

The Fig. 5 shows an example for the ability of the method to measure at different temperatures down to the phase transition of the investigated Potassium Dihydrogen Phosphate (KDP). The effects are low and remain low also near the phase transition contrary to the prediction of a Curie-Weiss law following the Landau theory. These results are discussed in another paper [9].

4. Conclusions

The presented stress apparatus together with the electronic setup allow precise and high resolution measurements of the nonlinear elastic properties of solids. The stress apparatus can be also used for other purposes like the measurement of electrostrictive coefficient when the dielectric constant is recorded via the applied stress. Further measurements are planned to confirm the results and to get information on the influence of dimension effects, sample preparation and heat treatment. These questions are especially important in the case of doped NaCl. Perhaps measurements down to liquid helium temperatures are of interest because of the growing importance of defects on the physical properties [10]. Cuts in 45°-direction of NaCl have to be also investigated to get the complete tensor of the nonlinear coefficients.

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