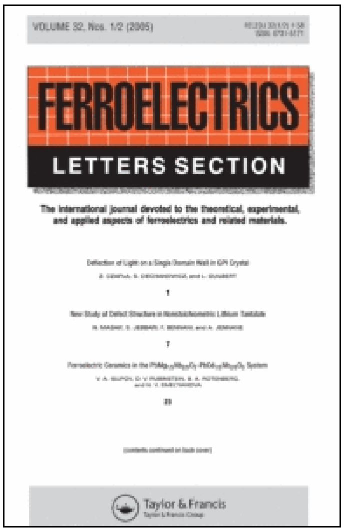

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OPTICAL SECOND HARMONIC MEASUREMENTS ON $KTaO_3$:Li †*

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Abstract Li^+ impurities replacing K^+ in $KTaO_3$ are known to induce a structural phase transition, the type of which yet is not very clear. Proposed models are a ferrodistortive transition of the lattice at 75 K [1], a polar glass state at low temperatures [2], or a paraelectric to ferroelectric transition in the impurity system below 50 K [3]. In order to test the different models we measured the optical second harmonic generation (SHG) efficiency of the crystal as a function of the sample temperature. The measurements favour the model of a paraelectric to ferroelectric transition in the impurity system.

Pure potassium tantalate ($KTaO_3$) is known to be an incipient ferroelectric as zero-point fluctuations suppress the ferroelectric phase transition [4]. Substitution of the A or the B site ion by smaller ions destabilizes the lattice significantly, and can induce a phase transition. Substituting the Ta ion by the smaller Nb on the B site is a rather effective possibility to induce ferroelectricity [5]. Less effective in order to destabilize the lattice is the substitution of K^+ by the smaller Na^+ on the A site which drives the ferroelectric transition, too [6]. The even smaller ion Li^+ was thought to have a similar destabilization effect as Na^+, but recently different models were proposed for the low-temperature phase of $KTaO_3$:Li: From acoustic

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resonance and from nuclear magnetic resonance measurements it was concluded, that the low temperature phase has no macroscopic spontaneous polarisation but is a so called 'polar glass' with fixed random-sited and randomly directed dipoles [7]. A second model suggested that the destabilisation by the Li-impurities drives a rotation of the TaO_6 octahedra which leads to a phase transition similar to the antiferrodistortive one in $SrTiO_3$ [1,8]. Yet birefringence measurements favoured the model of a ferroelectric transition with domains of fixed directed impurity dipoles [3]. By means of SHG measurements it should be possible to test the symmetry of the low temperature phase in order to determine the best model.

For the measurements a flux-grown $KTaO_3$ single crystal was used, which contained 3.5 % Li [9]. The sample size was about 5x3x3 mm^3, cut parallel to the cubic crystallographic faces. As fundamental light source for sample excitation served a Q-switched Nd^{3+}-YAG-Laser with a peak power of about 20 kW, pulse repitition rate of 1 kHz, and pulse length of about 300 nsec. The light was directed parallel to one of the cubic axes and polarised parallel to a second one by means of a prism polariser. The generated second harmonic light was separated from the fundamental wavelength by a suitable arrangement of filters and prisms and detected by an S-11 photomultiplier. Both relevant polarisation directions - parallel and perpendicular to the fundamental one - could be measured. The electronic detection system consisted of a gated photon counter and a microprocessor-controlled temperature measuring and data-acquisition system.

The measured SHG intensity (sum of both polarisations) is shown in Fig. 1. Due to the cubic symmetry in the high temperature phase of the crystal no SHG signal is found above the phase-transition temperature. Below this temperature a steep increase in the SHG signal is

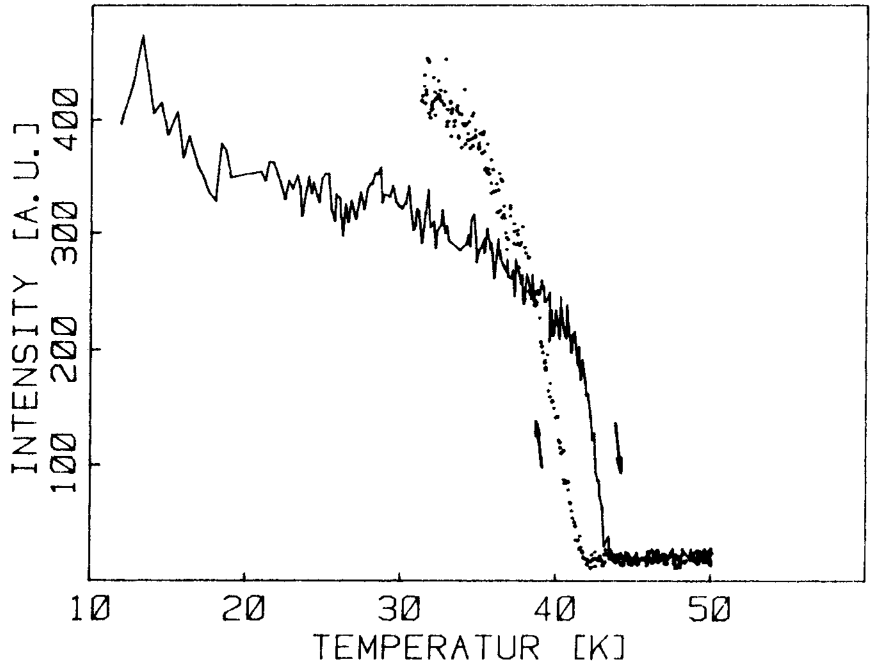

FIGURE 1 SHG-signal in $KTaO_3$:Li as a function of the sample temperature.

seen, which indicates that in the low temperature phase the crystal consists of polarised domains, the magnitude of which should be at least in the region of the wavelength of the light. If the crystal would exhibit an antiferrodistortive phase transition like $SrTiO_3$, one should find no or only an extremely weak SHG signal due to gradient terms [10,11], as the "normal" SHG signal would vanish because of the inversional symmetry of the distorted phase. Also, in the case of a polar glass state [2] one should find vanishing SHG intensity as the SHG signal in this case can be expressed as a sum over random-phased harmonic sources - i.e. the randomly directed impurity momenta - which would add up to zero.

The analysis of the polarised measurements shows an agreement with the model of a tetragonal low temperature phase in the impurity system with the nonlinear susceptibility tensor components $|d_{31}|=0.35\ |d_{33}|$. The thermal hysteresis of about 2.5 K is the same as found in birefringence measurements [3]. So the SHG measurements on $KTaO_3$:Li support the model of a first order paraelectric to ferroelectric phase transition in the impurity system.

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