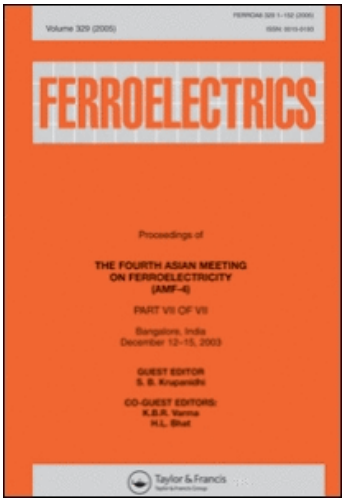

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Second harmonic generation near the phase transition of $SrTiO_3$

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SECOND HARMONIC GENERATION NEAR THE PHASE TRANSITION OF $SrTiO_3$

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Optical second harmonic generation (SHG) measurements were performed on $SrTiO_3$ crystals near the 107 K antiferrodistortive phase transition. Besides a temperature-independent SHG signal a strongly polarized peak in the SHG intensity near the phase transition was found. Furthermore our measurements show increasing SHG intensity towards lower temperatures below the phase transition.

INTRODUCTION

Optical second harmonic generation (SHG) measurements have been proved as a good tool to probe microscopic transformations of the crystal symmetry near phase transitions[1-3]. Pronounced effects on the SHG intensity can be seen when the crystal structure changes from centro- to noncentrosymmetric at the phase transition point[1]. In these transitions the lowest order nonlinearity is the dominating effect in SHG. To study higher order nonlinear processes we chose for our measurements $SrTiO_3$ as a crystal which is centrosymmetric both below and above the phase transition. The perovskite $SrTiO_3$ undergoes a second order antiferrodistortive phase transition at $T_o \approx 107$ K below which the TiO_6-octahedra alternately rotate around one of the cube axis. The temperature dependence of the rotation angle is given by $\langle\phi\rangle \propto (T_o - T)^{\beta}$ with $\beta = 0.34$[4,5]. In addition, $SrTiO_3$ exhibits high linear susceptibility which, according to Miller's rule, should cause a good efficiency for nonlinear effects.

EXPERIMENTAL

The measurements of the second harmonic intensity as function of temperature were performed using the experimental arrangement shown in Fig. 1. The light of a Q-switched Nd^{3+}-YAG-Laser with a repetition rate of 1 KHz, peak power of 20 kW, and pulse length of 300 nsec served as fundamental beam. The $SrTiO_3$ sample (typical size: 5x5x5 mm^3) was mounted in a liquid-nitrogen cooled cryostat, which allowed a temperature variation between 80 and 140 K. The SHG signal was detected by a photon-counting system consisting of a cooled S11-photomultiplier, a

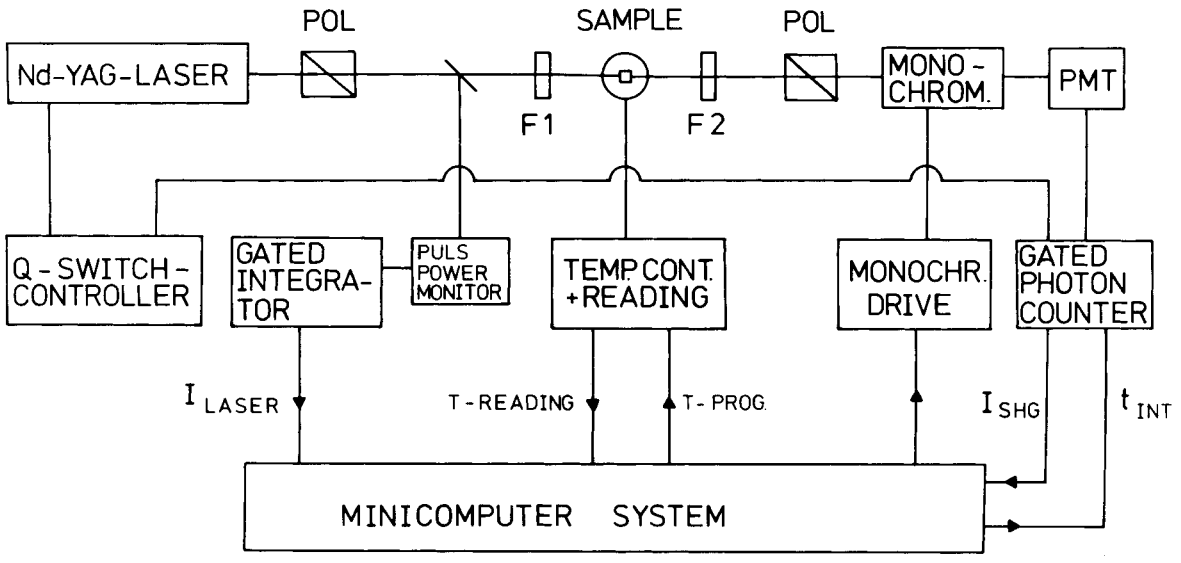

Figure 1: Experimental setup for measuring the temperature dependence of the SHG intensity in $SrTiO_3$. F1 and F2 are IR-edge (Schott RG 830) and IR-blocking filters (Schott BG 18), respectively.

gated photon counter, and a minicomputer. The measured SHG intensity was normalized to the square of the laser intensity. By means of two prism polarizers, the polarization of fundamental and second harmonic beam, respectively, could be defined.

RESULTS AND DISCUSSION

The measured temperature dependence of the SHG intensity is shown in Fig. 2 both for SHG polarization parallel and perpendicular to the incident polarization. Polarization and beam directions have been chosen parallel to the crystal axis, as no phase matching for SHG is possible in $SrTiO_3$. Total integration times per measured point of about 1000 seconds were used, the typical SHG intensities being 1 detected photon per second. Well above and below the phase transition the two curves show up the same features: Temperature independence above and additional intensity which increases to lower temperatures below the phase transition. In addition, the parallel polarized SHG signal exhibits a peak at the phase transition temperature having a width of about 1 K.

The constant background in the SHG intensity is probably due to impurities which are polar themselves as for instance OH^- [6] or lead to a microscopic distortion of the crystal symmetry. The increase in intensity to lower temperatures can be explained by higher order contributions to the SHG. In centrosymmetric crystals the lowest order non-vanishing term in the nonlinear polarization is a spatial gradient term which may be written as[7]

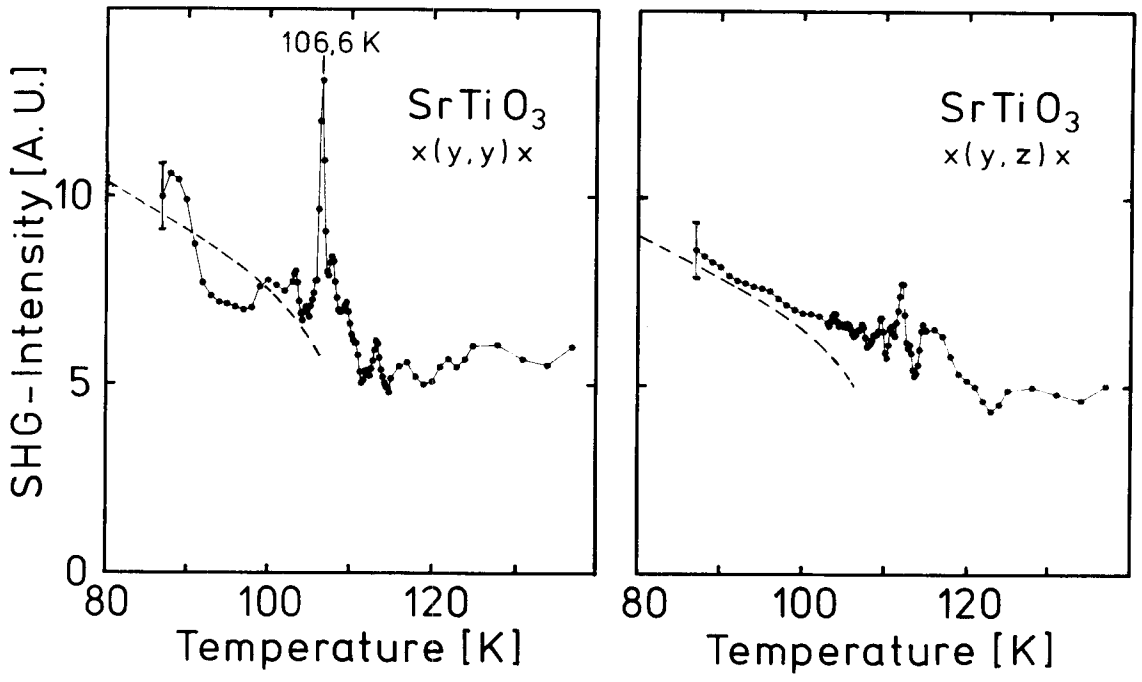

Figure 2: Temperature dependence of the SHG intensity in $SrTiO_3$, second harmonic polarization parallel (left) and perpendicular (right) to the fundamental beam polarization. The measured values are given by the dots, typical measurement errors are indicated by the bars.

$$P_2^{NL} = \chi^{NL} \cdot E_1 \cdot \nabla E_1$$

where χ^{NL} is a fourth rank tensor, and E_1 and ∇E_1 are the macroscopic electric field and its spatial gradient, respectively. In $SrTiO_3$ χ^{NL} should vanish in the cubic phase[8] and be proportional to the rotation angle of the TiO_6 octahedra in the tetragonal phase below the phase transition temperature. As the rotation angle $\langle\phi\rangle \propto (T_o-T)^{\beta}$ and the additional SHG intensity $I_2 \propto (P_2^{NL})^2$, the temperature dependence of I_2 can be written as

$$I_2 \propto (T_o-T)^{2\beta}.$$

This dependence is given by the dashed curves in Fig. 2. A comparison to KDP yields as numerical value for the nonlinear susceptibility due to the spatial gradient (using that $|\nabla E| = |k| \cdot |E|$ for a plane wave):

$$k \cdot \chi^{NL}_{ijkl}(SrTiO_3) \approx 10^{-4} \cdot \chi_{36}(KDP) \cdot [T_o-T)/T_o]^{\beta}.$$

This value is in reasonable agreement with values measured by phase matched SHG in other centrosymmetric materials[7,8].

The SHG peak at the phase transition temperature only shows up in

the measurements with parallel polarization of fundamental and harmonic wave; that means that in the nonlinear susceptibility tensor responsible for this peak the diagonal elements are at least 3 times the value of the off-diagonal elements. This would explain that the peak in the perpendicular polarization is at least one order of magnitude smaller and so cannot be resolved. Responsible for this peak should be one of the mechanisms proposed to explain central peaks[9] as they are found for instance in light scattering experiments:

a) Intrinsic mechanisms as for instance fluctuation effects or

b) Extrinsic mechanisms as for instance defect motion or ordering or changes in domain structure around the phase transition.

All of these mechanisms cause local distortions of the inversional symmetry of the crystal near the phase transition temperature and so increase the nonlinear susceptibility of the crystal.

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