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学位論文題目	

**Ferroelectric phase transition and dynamical behavior of PNRs of potassium tantalate niobate crystals : broadband inelastic light scattering study**

(広帯域非弾性光散乱分光法によるタンタルニオブ酸カリウム結晶の強誘電相転移とナノ極性領域のダイナミクスの研究)

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## 論 文 の 要 旨

This dissertation is devoted to clarify the dynamical behaviors of polar nanoregions (PNRs) in high quality Nb rich  $\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$  (KTN) and Li-doped KTN single crystals by inelastic light scattering. The dielectric measurements were also carried out to understand the complex dynamical behavior of relaxor-like ferroelectrics.

The precursor dynamics of relaxor-like ferroelectric phase transitions in non-doped KTN and Li-doped KTN single crystals were investigated by Brillouin and Raman scatterings. In the vicinity of the Curie temperature,  $T_C$ , the remarkable softening of the sound velocity and increase in the sound attenuation were observed in both non-doped and Li-doped KTN single crystals. The increase in the central peak (CP) intensity and sound attenuation as well as significant softening of the VTA were clearly observed below the intermediate temperature,  $T^*$ , indicating the start of the rapid growth of the dynamic PNRs. The temperature variation of the size of a dynamic PNR in both crystals is evaluated, and below the  $T^*$ , it markedly increases down to  $T_C$  towards the percolation limit. In non-doped and Li-doped KTN, the relaxation time estimated by the width of a broad CP shows the critical slowing down towards  $T_C$ , which is the evidence of the order-disorder nature of the ferroelectric phase transition.

The Li-doping effect on KTN crystals was clearly seen as the stretching of the slowing down and broadening of the elastic anomaly in the 5%Li-doped KTN in comparison with that of the non-doped KTN by Brillouin scattering. The enlargement of the temperature region of the elastic anomaly in Li-doped KTN can be attributed to the fact that the random fields by doped Li

ions enhance the growth of PNRs. By the Li-doping, the remarkable increase of the size of the dynamic PNRs towards TC was also found. The broadening of the elastic anomaly can be the evidence of the enhanced relaxor nature of 5%Li-doped KTN single crystals. The enhanced relaxor nature of KTN by the Li-doping was also confirmed by the dielectric properties using the extended Curie-Weiss law. In the 5%Li-doping in KTN, the transformation of symmetry from  $A1(z)$  to  $E(x,y)$  of the local modes in PNRs was observed by the angular dependence of Raman scattering. The first-principles calculation shows the partial occupancy of Li ions at B-site. The total energy at A- and B-site occupancy of the perovskite structure for the Li-doped KTN was calculated by the density functional theory. In a stable equilibrium state, 4.2%Li ions occupy A-site, while the rest of 0.8% Li ions occupy B-site. Thus, the increase of the random field strength induced the partial occupancy of Li ions at B-site can be caused by the hetero-valence charge disorder at B-site. Such an enhancement of random fields can be the physical origin on the suppression of the critical slowing down of the relaxation time near TC and the change of the local symmetry of PNRs.

## 審 査 の 要 旨

〔批評〕

圧電材料や電気光学材料として重要な強誘電体の産業応用の大半はペロフスカイト構造酸化物である。本論文は、キュリー温度直上における巨大電気光学効果や巨大電歪効果などの巨大な外場応答で注目されている非鉛系リラクサー強誘電体KTNについて、ナノ極性領域に関連する弾性的性質を非破壊・非接触で測れる顕微ブリルアン散乱法、並びに高分解能ラマン分光法を用いて、その強誘電相転移のダイナミクスを初めて明らかにした独創性と新規性の高い優れた研究である。上述したこれらの研究成果は、ペロフスカイト構造の非鉛系リラクサー強誘電体の外場応答やその強誘電相転移のダイナミクスを実験的に捕らえた重要な研究であり、巨大電場応答を示すリラクサー強誘電体の物性理解のみならず、磁性を含むマルチフェロ物質などへの波及効果など今後の物質工学、固体物理学等の進歩に大きく貢献すると判断される。

〔最終試験結果〕

平成 28 年 2 月 19 日、数理物質科学研究科学学位論文審査委員会において審査委員の全員出席のもと、著者に論文について説明を求め、関連事項につき質疑応答を行った。その結果、審査委員全員によって、合格と判定された。

結論〕

上記の論文審査ならびに最終試験の結果に基づき、著者は博士(工学)の学位を受けるに十分な資格を有するものと認める。