

## **FINAL REPORT, GRANT DE-FG02-06ER46318, PI : J. TOULOUSE**

“Multiscale Dynamics of Relaxor Ferroelectrics”

### **PAPERS**

- A new scattering mechanism of acoustic phonons in relaxor ferroelectrics: the case of  $\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$ , J. Toulouse, E. Iolin, B. Hennion, D. Petitgrand, G. Yong, R. Erwin, arXiv:1001.4096
- Dielectric Transparency induced by Hetero-Phase Oscillations in Relaxor Ferroelectrics, Jean Toulouse, R.K. Pattnaik, L.A. Boatner, arXiv:1203.2639
- Theoretical Study of the Soft Optic Mode Dynamics in a Relaxor Ferroelectric: the Effect of Polar Nanoregions , E. Iolin, J. Toulouse, arXiv:1007.3537
- The Three Characteristic Temperatures of Relaxor Dynamics and their Meanings Ferroelectrics, J. Toulouse, Ferroelectrics, 369; 203-213 (2008).

### **PRESENTATIONS**

#### **2012**

- Critical Slowing Down in the Relaxor Ferroelectric  $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ , L. Cai and J. Toulouse, APS March 2012 meeting Boston (MA), B32-6
- EIT-like Effect due to Hetero-Phase Oscillations near the Phase Transition of Relaxor Ferroelectrics, J. Toulouse, R.K. Pattnaik, L.A. Boatner, APS March 2012 meeting Boston (MA), B32-7
- Temperature Evolution of the Linear Birefringence in Striated Single Crystals of KTN D. Jackson, R.K. Pattnaik, A. Agranat and J. Toulouse, APS March 2012 meeting Boston (MA), B32-8
- Temperature Evolution of the Linear Birefringence in Striated Single Crystals of KTN D.Jackson, R.K. Pattnaik, A. Agranat and J. Toulouse , Workshop on Fundamental Physics of Ferroelectrics and Related Materials, Argonne National Laboratory (Jan 2012), refereed extended abstract

#### **2011**

- Comparison of the Raman Low Frequency Spectra of NBT and KLT, D. Jackson, R.K.Pattnaik, D. Viehland, H. Luo and J. Toulouse, American Physical Society March meeting (2011), Dallas (TX), L33-4

#### **2010**

- Understanding NBT through a comparison with KTN and KLT, J. Toulouse, International Workshop on Lead Free Ferroelectrics Subject, Shanghai, China, August 2010 (invited)
- Resonance scattering of the transverse optic mode by polarized nano regions, E. Iolin and J. Toulouse, Advances in the Fundamental Physics of Ferroelectrics and Related Material, Feb.2010, Aspen (CO), refereed extended abstract

#### **2009**

--Critical Oscillation-Induced Dielectric Transparency in the Relaxor Ferroelectric  $K_{1-x}Li_xTaO_3$ , J. Toulouse, International Meeting on Ferroelectricity, Xi'an (China), August 2009, refereed conference paper

--The Three Characteristic Temperatures of Relaxor Dynamics and their Meaning, J. Toulouse, APS March 2009 meeting, Pittsburgh (PA) T-41-8

--Scattering of the Transverse Acoustic Phonon by Polar NanoDomains in the Relaxor Ferroelectric  $KTa_{1-x}Nb_xO_3$  (KTN), J. Toulouse and E. Iolin, APS March 2009 meeting, Pittsburgh (PA), S1-69

## 2008

--Transverse Acoustic Mode Dynamics in the Relaxor of  $KTa_{1-x}Nb_xO_3$  (KTN), J. Toulouse, E. Iolin, B. Hennion, D. Petitgrand, American Conference on Neutron Scattering, Santa Fe (NM) May 2008 and American Conference on Neutron Scattering June 2008, Lund Sweden, refereed conference paper

## 2007

--Neutron Scattering Study of the Phase Transition(s) in the Mixed Ferroelectric single Crystal,  $KTa_{1-x}Nb_xO_3$  (KTN), E. Iolin and J. Toulouse, European Neutron Society Annual Meeting, Lund, Sweden (July 2007) and Williamsburg Conference on Ferroelectrics, Williamsburg (VA) Feb. 2007

--The Three Characteristic Temperatures of Relaxor Dynamics and their Meanings, J. Toulouse, 11th European Meeting on Ferroelectricity, EMF11, Bled, Slovenia, September 2007, refereed conference paper

## NARRATIVE

The purpose of this project was to better understand the "Multiscale Dynamics of Relaxor Ferroelectrics". The output of the project is detailed above. The results of the work were presented at a number of different conferences and four papers were written.

The multiscale dynamics of relaxors was clearly identified in the three characteristic temperatures that were identified. In particular, we were the first group to identify an intermediate temperature,  $T^*$ , at which the correlations between off-center ions in relaxor cross-over from being dynamic to being static and giving rise to the characteristic relaxor behavior in the dielectric constant. Other groups have now confirmed the existence of such an intermediate temperature. (see e.g. [E. Dul'kin](#), [S. Kojima](#) and [M. Roth](#) Euro. Phys. Lett. 97 5 57004, 2012)

We also made and reported two other observations:

- i) a coherent interference phenomena (EIT-like effect) near the transition of several relaxors, which provides information on the nature and mechanism of the transition. Deeming this result important, we submitted it to Physical Review Letters. After a lengthy process, the paper was not accepted. Without changing the essence of the paper, the interpretation is now being recast in a more relevant form in the framework of a Fano-type resonance. Simultaneously, we are running more experiments in order to strengthen the interpretation. The paper will be resubmitted in the next few months either to Phys. Rev. Lett or to Euro. Phys. Lett. or in an expanded form to PRB

This work has also led us to discover the essential impact of the spatial distribution of the off-center ions on the macroscopic properties of relaxors. New work is ongoing on this particular aspect.

- ii) in a similar way, inelastic neutron scattering results were interpreted as resonant scattering of acoustic phonons by localized modes in polar nanodomains. Also submitted to PRL, the paper was also not accepted after a lengthy review/response process. Given the size of polar nanoregions (1-10 nm) and the observation of localized modes around much smaller disturbances/defects in other solids, such localized modes must exist and their scattering of acoustic phonons must be an important characteristic of relaxors and we are again re-examining our neutron data to strengthen the argument in view of a resubmission, probably to Phys. Rev. B.

In parallel with the neutron scattering work, we also developed a theory of the scattering of phonons by the above localized modes. The theoretical development is very formal at this point and did not allow an easy comparison with the experimental results. This work is in progress.

As indicated above, the three papers in revision are posted on the database arXiv.

In addition, we have obtained a contract from Cambridge University Press to write a book on "Disordered Ferroelectrics". This book is in progress with delivery of the manuscript scheduled for August 2013.