Phonons in $SrTiO_3$ and $EuTiO_3$ investigated with neutron scattering

Olivier Delaire[1], Jiawang Hong[1], Huibo Cao[2], Jiaqiang Yan[1],

Barry Winn[2], Songxue Chi[2], G. Ehlers[1], Lynn Boatner[1], Brian Sales[1]

[1]Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN, USA
[2]Quantum Condensed Matter Division, Oak Ridge National Laboratory, Oak Ridge, TN, USA

$SrTiO_3$ (STO) is an incipient ferroelectric material, whose phonons have been studied in seminal works [1-5]. While the zone-center transverse optic (TO) mode softens with decreasing temperature, the ferroelectric transition is suppressed by quantum fluctuations [6]. Another distortion, corresponding to alternative tilts of oxygen octahedra, causes a stronger instability and result in a transition from cubic perovskite to an antiferrodistortive (AFD) tetragonal phase at Tc=105K. The AFD transition involves the softening of the R_{25} phonon mode at the R-point. The temperature dependence of both the zone-center TO and the R_{25} mode can be understood in principle on the basis of anharmonic interatomic potentials. However, a more detailed understanding of anharmonic effects is still lacking, in particular for the renormalization of phonon frequencies at finite temperature. In addition, recent re-examination of the dynamics of STO have emphasized the competition between FE and AFD instabilities [7], but a full understanding of coupled instabilities has not yet emerged.

$EuTiO_3$ (ETO) presents a number of similarities with STO, with the cubic to tetragonal AFD distortion occurring at Tc=280K [8,9]. ETO is also unique owing to the presence of magnetic Eu ions. By applying a tuning parameter, such as bi-axial tension, ETO can be turned into a ferroelectric ferromagnet, the ideal multiferroic. [10] However, in unstrained ETO, the ferroelectric TO mode does not exhibit as much softening as in STO, and its frequency remains above ~10meV at all temperatures, precluding the formation of the desired ferroelectric state. The origin of this behavior remains an open question, and both quantum fluctuations or spin-phonon interactions could be involved.

We will present results of extensive lattice dynamics measurements in single crystals with inelastic neutron scattering (INS), on single crystals of STO and ETO (synthesized with ^{153}Eu), highlighting similarities and differences. In addition, we present results of first-principles computations of the dynamical structure factor, $S(\mathbf{Q},E)$, including anharmonic effects stabilizing the cubic phase at finite temperature in both STO and ETO.

Acknowledgements We acknowledge funding from the DOE Office of Science, Basic Energy Science.

References

[1] R. A. Cowley, Phys. Rev. **134**, A981 (1964).
[2] R. A. Cowley, *et al.*, Solid St. Commun. **7**, 181 (1969)
[3] G. Shirane and Y. Yamada, Phys. Rev. **177**, 858 (1969)
[4] S. M. Shapiro, J. D. Axe, G. Shirane, and T. Riste, Phys. Rev. B **6**, 4332–4341 (1972).
[5] P. A. Fleury and J. M. Worlock, Phys. Rev. **174**, 613 (1968)
[6] K. A. Müller and H. Burkard, Phys. Rev. B **19**, 3593 (1979)
[7] U. Aschauer and N. Spaldin, J. Phys.: Condens. Matter **26**, 122203 (2014)
[8] D. S. Ellis *et al.*, Phys. Rev. B **86**, 220301(R) (2012)
[9] A. Bussmann-Holder *et al.*, Phys. Rev. B **83**, 212102 (2011)
[10] J. H. Lee, et al., *Nature* **466**, 954 (2010).