Exotic Caloric Effects Predicted from First-principles Simulations

I. Ponomareva[1,*] and S. Lisenkov[1]
[1]Department of Physics, University of South Florida
4202 E. Fowler Ave, Tampa, FL, 33620
*Inna Ponomareva: iponomar@usf.edu

First-principles-based simulations play an important role in the exploration of caloric effects in complex ferroics. For example, the predictions of elastocaloric and multicaloric effects in classic ferroelectrics [1,2] promoted experimental demonstrations of such effects in ferroelectric ceramics [3,4]. In this talk I will focus on our recent developments of atomistic simulation tools that led to the prediction of some exotic caloric effects.

One example will be the caloric effects in antiferroelectrics. Such materials are very appealing for electrocaloric effect investigation since they do not possess spontaneous polarization but yet could exhibit dielectric constants comparable to ferroelectrics. We combined thermodynamic approach with first-principles simulations to predict the existence of a scaling law for the electrocaloric temperature change in antiferroelectrics. Another interesting feature of antiferroelectrics is their ability to exhibit strong electromechanical coupling which could potentially lead to the existence of large piezocaloric effect. Such an effect describes the adiabatic change in temperature in response to the application of the stress field. We used our first-principles-based simulations to predict the existence of highly tunable piezocaloric effect in antiferroelectric $PbZrO_3$. One of the interesting findings is the linearity of electrocaloric change in temperature in applied stress which could be used to double the efficiency of solid state refrigeration cycle. Interestingly, it appears that the nanoscale antiferroelectrics could have even more surprises in stock. It turns out that the competition between the ferroelectric and antiferroelectric phases that is characteristic of antiferroelectrics could be greatly changed at the nanoscalee. In particular, the ferroelectric phase could be stabilized in really thin antiferroelectric samples. We used atomistic simulations to explore potential of such films to exhibit electrocaloric effect. Our simulations predict that the antiferroelectric-ferroelectric phase competition leads to the enhancement of the electrocaloric effect along with the broadening of the electrocaloric response. Next I will turn to rather unexpected predictions from atomistic simulations. One example is the flexocaloric effect in ferroelectric ultrathin films. In such an effect the change in temperature is caused by the adiabatic application of strain or stress gradient. Another example is the prediction of surprisingly large elastocaloric effect in materials that are not members of ferroic family. In particular, the elastocaloric effect of up to 30 K is predicted in carbon nanotubes and graphene from atomistic simulations.

1. S. Lisenkov and I. Ponomareva Phys. Rev. B **86**, 104103 (2012).
2. S. Lisenkov, et. al. Phys. Rev. B **87**, 224101 (2013).
3. A. Chauhan, S. Patel, and R. Vaish, Appl. Phys. Lett. **106**, 172901 (2015).
4. A. Chauhan, S. Patel, and R. Vaish, Acta Mater. **89**, 384 (2015).