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Flexoelectric properties of ferroelectrics and the nanoindentation size-effect

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ABSTRACT

Recent works have established the critical role of flexoelectricity in a variety of size-dependent physical phenomena related to ferroelectrics including giant piezoelectricity at the nanoscale, dead-layer effect in nanocapacitors, dielectric properties of nanostructures among others. Flexoelectricity couples strain gradients to polarization in both ordinary and piezoelectric dielectrics. Relatively few experimental works exist that have determined flexoelectric properties and they all generally involve some sort of bending tests on micro-specimens. In this work, we present a straightforward method based on nanoindentation that allows the evaluation of flexoelectric properties in a facile manner. The key contribution is the development of an analytical model that, in conjunction with indentation load–displacement data, allows an estimate of the flexoelectric constants. In particular, we confirm the experimental results of other groups on $BaTiO_3$ which differ by three orders of magnitude from atomistic predictions. Our analytical model predicts (duly confirmed by our experiments) a strong indentation size-effect due to flexoelectricity.

1. Introduction

Piezoelectricity exists only in non-centrosymmetric crystals. However, a somewhat under-appreciated fact is that all dielectrics polarize when subjected to inhomogeneous strain. This phenomenon, the coupling of strain gradients to polarization, is known as flexoelectricity. Phenomenologically, the total polarization can be expressed as:

$$P_i = \underbrace{d_{ijk}\varepsilon_{jk}}_{=0,\ for\ centrosymmetric\ materials} + \mu_{ijkl}\frac{\partial \varepsilon_{jk}}{\partial x_l}. \quad (1)$$

Recently, flexoelectricity has generated much excitement due to the elucidation of several insights relevant at the nanoscale. For example, Catalan et al. (2004) have studied the impact of flexoelectricity on the dielectric properties and Curie temperature of ferroelectric materials while Cross and coworkers (1999, 2006) have proposed tantalizing notions such as "piezoelectric composites without using piezoelectric materials". Eliseev et al. (2009) have investigated the renormalization of properties of ferroelectric nanostructures due to the spontaneous flexoelectric effect as well as developed analytical approaches to derive size-effects in such nanostructures (Eliseev and Morozovska, 2009). One of us has demonstrated strong size-dependent enhancement of the apparent piezoelectric coefficient in materials that are intrinsically piezoelectric (Majdoub et al., 2008a, 2009b) as well as explored ramifications for energy harvesting (Majdoub et al., 2008b, 2009c). More recently Majdoub et al. (2009a) have also demonstrated, through first principles and theoretical calculations, that the so-called dead-layer effect in nanocapacitors may be strongly influenced by flexoelectricity. The reader is referred to reviews by Tagantsev (1986, 1991), Tagantsev et al. (2009) for further details.

Relatively few experimental works exists on the determination of flexoelectric properties of crystals. Cross and co-worker's pioneering work provided some of the first data on various perovskites like PMN, PZT, BST, and $BaTO_3$ (Ma and Cross, 2001, 2002, 2003, 2006; Fu et al., 2006, 2007). More, recently Zubko et al. (2007) have published the experimental characterization of the complete flexoelectric tensor for $SrTiO_3$. The afore-mentioned experimental approaches are predicated on bending experiments and are decidedly non-trivial. In parallel, various groups have also made atomistic predictions of flexoelectric properties. For example, one of us (Maranganti and Sharma, 2009) presented results for a number of dielectrics of technological and scientific interest. Dumitrica et al. (2002), Kalinin and Meunier (2008) discuss graphene and very recently, Hong et al. (2010) presented a first principles approach and consequent data for both $SrTiO_3$ and $BaTO_3$. While the theoretical works of various groups are all in agreement, the experimentally estimated flexoelectric constant of $BaTO_3$ is 3 orders of magnitude higher compared to the atomistically predicted value. The reasons for this discrepancy are still an open research issue (and beyond the scope of the present paper).

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