**Nanostructured multiferroic Pb(Zr,Ti)O3−NiFe2O4 thin-film composites**

Aleksander Matavž,1 Primož Koželj,1 Maximilian Winkler,2 Korbinian Geirhos,2

Peter Lunkenheimer,2 and Vid Bobnar1\*

1Condensed Matter Physics Dept., Jožef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia

2Experimental Physics V, Center for Electronic Correlations and Magnetism, University of Augsburg, 86159 Augsburg, Germany

\*E-mail: vid.bobnar@ijs.si

The synthesis of multiferroic composite materials rises as an alternative method for simultaneously obtaining ferroelectric and ferromagnetic properties at room temperature with a high degree of magnetoelectric coupling. Compared to bulk composites, thin-films exhibit unique advantages: their composition and connectivity can be modulated at the microscopic scale, which eventually leads to the artificial heterostructures with huge application potential in novel functional devices in the fields of sensors, data storage, and energy harvesters. In composites of spinel ferrites and ferroelectric ceramics − they have up to now been prepared as bulk ceramics or in thin-film configuration as bi- or multi-layers − the magnetoelectric effect arises from direct stress coupling between magnetostrictive (ferromagnetic) and piezoelectric (ferroelectric) grains. It is thus evident that good and extensive connectivity among the constituents might result in significant coupling between piezoelectric and magnetostrictive phases and enhance the magnetoelectric effects.

We present multiferroic thin-film composites with a novel morphology that in fact provides such an extensive connectivity between the magnetostrictive and piezoelectric constituents. They were fabricated by embedding the ferromagnetic NiFe2O4 into self-assembled highly-porous ferroelectric Pb(Zr,Ti)O3 thin films (the latter themselves exhibit extremely interesting functional properties − the porosity-mediated release of the substrate’s mechanical constraints namely boosts their piezoelectric response to the level of bulk ceramics [1]). Detailed structural investigations of developed composites revealed a two phase pure system, without any chemical reaction or solid solubility between both constituents during synthesis. The multiferroicity is clearly evidenced by detecting both ferroelectric and ferromagnetic hysteresis loops at room temperature. Detected magnetic field-induced changes of the dielectric constant, not only at low frequencies but also above the characteristic frequency of the Maxwell-Wagner behavior, reveal a direct stress coupling between the magnetostrictive NiFe2O4 and piezoelectric Pb(Zr,Ti)O3 grains and imply a great potential utility of the developed material in magnetocapacitive applications [2].

Keywords: Nanostructured films, Thin-film composites, Multiferroics, Magnetoelectrics.

[1] A. Matavž, A. Bradeško, T. Rojac, B. Malič, V. Bobnar, Self-assembled porous ferroelectric thin films with a greatly enhanced piezoelectric response, Applied Materials Today 16, 83 (2019).

[2] A. Matavž, P. Koželj, M. Winkler, K. Geirhos, P. Lunkenheimer, V. Bobnar, Nanostructured multiferroic Pb(Zr,Ti)O3–NiFe2O4 thin-film composites, Thin Solid Films 732, 138740 (2021).