



	<b>Experiment title:</b> Strain Profile of Magnetoelectric Interfaces by X-ray Diffraction Methods	<b>Experiment number:</b> 28-01-1036
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We investigated the mechanical coupling in magnetoelectric (ME) epitaxial composites between a  $\text{CoFe}_2\text{O}_4$  (CFO) magnetostrictive single crystal substrate and a  $\text{PbZr}_{0.52}\text{Ti}_{0.48}\text{O}_3$  (PZT) piezoelectric thin film. The coupling was determined by measuring the lattice deformation in the both materials under an applied magnetic field. Single crystals of CFO were prepared by the borax flux technique [1] which have octahedron shapes with (111) facets. An epitaxial PZT layer of 200 nm thickness is grown on the top of one of (111) surfaces via pulsed-laser deposition. X-ray diffraction (XRD) experiments were carried out at the BM28 beamline at an energy of 15 keV. Using a Si (111) single crystal analyzer positioned between the detector and the sample a very high  $q$ -resolution was obtained. Figure 1 shows {400} and {333} cubic Bragg reflections of CFO in the CFO/PZT composite. The {400} and {333} reflections show four and two peaks, respectively. The peaks were separately fitted with gaussian functions. The intensity ratio is 2:2:1:1 for {400} and 1:1 for {333}. These features show the presence of two domains with the same symmetry and the same proportion. The {400} cubic reflection splits into 4 CFO peaks,  $\mathbf{a}_1$ ,  $\mathbf{a}_2$ ,  $\mathbf{c}_1$  and  $\mathbf{c}_2$ , showing that CFO switches to a lower symmetry. In the new tetragonal phase the proportion of  $\mathbf{a}_1$  and  $\mathbf{a}_2$  domains is two times higher than that of the  $\mathbf{c}_1$  and  $\mathbf{c}_2$  domains.

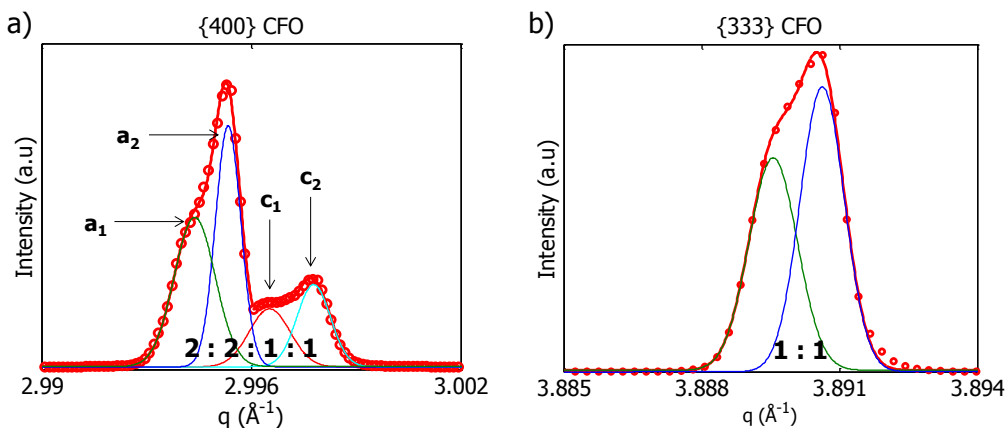


Figure 1: (a) {400} and b) {333} CFO Bragg reflections. The splittings of {400} and {333} cubic reflections reveal the tetragonal structure with the existence of 2 domains in CFO.

Figure 2 shows {400} CFO and {200} PZT Bragg reflections in the CFO/PZT composite in the presence of an external magnetic field applied along [100] direction as seen in the inset of Fig. 2c. While the shift of the Bragg peak positions is only visible in PZT, the magnetic field diminishes the intensity of  $\mathbf{a}_1$  and  $\mathbf{a}_2$  peaks and enhances the intensity of  $\mathbf{c}_1$  and  $\mathbf{c}_2$  peaks. The changes in intensity can be explained by two domains switchings which take place at two different values of the magnetic field. The first starts at  $H=0$  when  $\mathbf{a}$  domains switch to  $\mathbf{c}$  domains and the second starts at  $H=2.4$  kOe when  $\mathbf{c}_1$  domains switch to  $\mathbf{c}_2$  domains. In taking in account domains switchings and using growth functions for describing the proportion of domains for each magnetic field, the data have been very well reproduced as shown in Fig. 3a. The position and the intensity of peaks can be used to calculate the field induced strain via  $\epsilon =$

$\epsilon \cdot (I_c(H) - I_c(H=0)) / (I_a(H) + I_c(H))$ , where  $\epsilon$  is the lattice distortion. Here,  $I_a$  and  $I_c$  are the integrated intensities of **a** and **c** domains, respectively. Fig. 3b shows the strain in the two domains of CFO and in PZT. A compressive strain is observed in the both materials with a maximum value of  $-6 \cdot 10^{-4}$  in CFO in agreement with literature values [2] and fits with the simple model explained above. The comparison of the strain in both materials reveals that the strain in the PZT layer is significantly lower than the strain in the CFO substrate. This means, that the transfer of the strain is not perfect at the interface between the two materials. The mechanical coupling is given by  $k = d_{\text{PZT}}/d_{\text{CFO}}$  where  $d_{\text{PZT}}$  and  $d_{\text{CFO}}$  are the magnetostriction coefficients of CFO and PZT, respectively, deduced from the slope of the strain versus magnetic field. This gives a weak coupling of only  $k = 0.6 \pm 0.1$ , a value close to that found in CFO/PMNPT epitaxial composites [3]. This is an unexpected result for epitaxial composite systems in which the coupling is supposed perfect. This is attributed to the grain boundaries which play an important role in the strain relaxation of crystallites and thus reduce the strain transfer at the interface between both components [3].

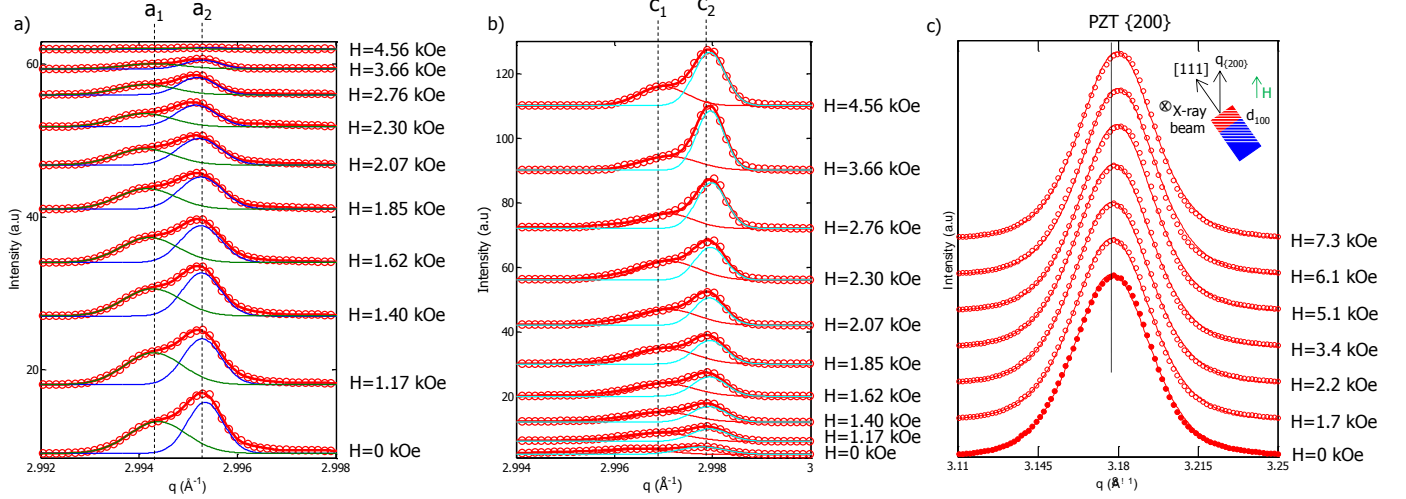


Figure 2: (a) (400)/(040), b) (004) CFO and (c) (200) PZT Bragg peaks in the CFO/PZT composite, measured during the application of an external magnetic field. The inset shows the X-ray experimental geometry.

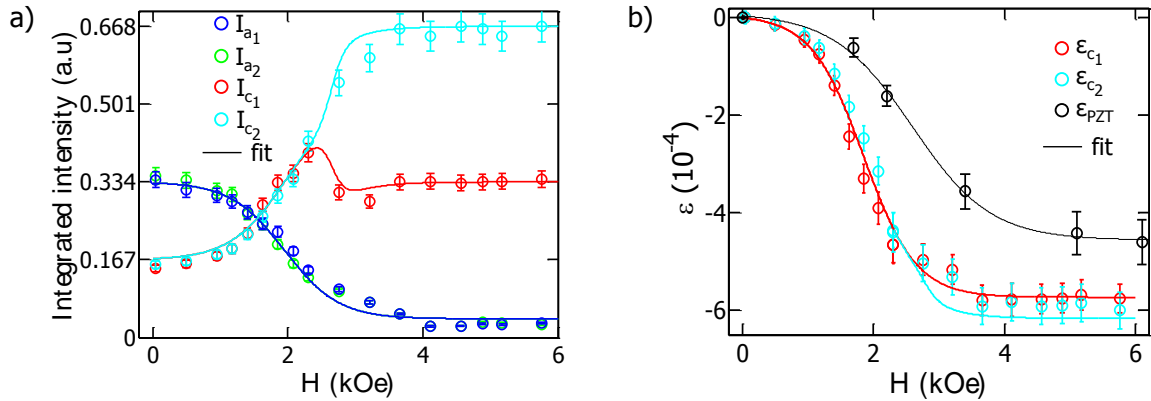


Figure 3: (a) Plot of magnetic field induced intensities corresponding to two CFO domains oriented to **a** and **c** axis. (b) Plot of magnetic field induced strains of the two domains in CFO and PZT. The solid lines show the fits deduced from the simple model.

In conclusion, to investigate the ME coupling at the CFO/PZT interface, we performed the magnetic field induced strain coupling in the CFO/PZT epitaxial composites at BM28. A value of saturation strain of  $6 \cdot 10^{-4}$  is observed in CFO in agreement with literature values. The study revealed that the magnetic field induced strain coupling is surprising low at the CFO/PZT interface. This is attributed to the grain boundaries which play an important role in the strain relaxation of crystallites.

In future work we plan to reproduce these promising results and perform systematic studies of related systems by this method.

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[1] N. Hur et al, Journal of Crystal Growth, 340, 171 (2012)

[2] R. Bozorth et al, Phys. Rev. 99, 1788 (1955)

[3] C. T. Koops, M. Abes, S. B. Hrkac, A. Petraru, H. Kohlstedt, O. H. Seeck, O. M. Magnussen, and B. M. Murphy, in preparation