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Magnetoelastic coupling in epitaxial cobalt ferrite/barium titanate heterostructures

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ABSTRACT

Ultra-thin cobalt ferrite films have been synthesised on ferroelectric barium titanate crystals. The cobalt ferrite films exhibit a magnetic response to strain induced by structural changes in the barium titanate substrate, suggesting a pathway to multiferroic coupling. These structural changes are achieved by heating through the phase transition temperatures of barium titanate. In addition the ferromagnetic signal of the substrate itself is taken into account, addressing the influence of impurities or defects in the substrate. The cobalt ferrite/barium titanate heterostructure is a suitable oxidic platform for future magnetoelectric applications with an established ferroelectric substrate and widely tuneable magnetic properties by changing the transition metal in the ferrite film.

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1. Introduction

Multiferroics are a field of intense and still increasing research interest [1–5]. These materials exhibit several ferroic ordering phenomena, thus, allowing simultaneous switching of multiple ferroic properties, e.g. an electrical switching of the polarisation with a simultaneous switching of the magnetisation. They promise exciting new applications in electronics and adjacent fields. Because of the fundamental requirements for the occurrence of ferroelectricity and ferromagnetism in a material the number of single phase multiferroic systems is limited [1,2,6]. Ferromagnetism typically requires the existence of partially filled subshells, e.g. a partially filled *d*-shell in transition metals. On the other hand most classic ferroelectrics like barium titanate consist only of atoms with a *d*⁰ configuration. Combining two ferroic systems in an epitaxial system via their interface is a popular approach to circumvent these limitations [1,2].

A well discussed mechanism for coupling of two ferroic systems is strain [7,8]. When a ferroelectric material is epitaxially connected to a magnetostrictive material the electrical and magnetic information can be conveyed as interface strain from one to the other. Because of its large magnetostrictive coefficient cobalt ferrite is an ideal material for these systems [7]. Barium titanate on the other hand is a well-established ferroelectric [8].

An advantage of barium titanate is that strain can also be induced by heating or cooling through its phase transitions. For this matter it exhibits three useful phase transitions: from rhombohedral to orthorhombic at 189 K, from orthorhombic to tetragonal at 285 K and from tetragonal to cubic at 407 K [8]. Of these three transitions we used the ones from rhombohedral to orthorhombic and orthorhombic to tetragonal in our investigation.

In this paper we show the magnetoelastic coupling of a thin cobalt ferrite layer to a barium titanate bulk substrate. To emphasise the important interface region of these systems in our measurements we use ultra-thin cobalt ferrite layers. Furthermore we take the ferromagnetic behaviour of the barium titanate substrate itself into account. This is necessary due to the ubiquity of ferromagnetic signals in diamagnetic oxide crystals as suggested by Khalid et al. [9]. Sources of these ferromagnetic signals may either be defects or impurities in the oxide crystals [9]. As these magnetic defect sites and impurities also experience the strain of the barium titanate crystal their magnetostrictive response cannot be neglected.

2. Methods

The BaTiO₃(001) crystals were pre-treated in an oxygen atmosphere of 1.0×10^{-6} mbar at 700 K. XPS measurements showed no contaminations with carbon or other elements. PLD was subsequently carried out in a 1.0×10^{-4} mbar atmosphere of oxygen at a sample temperature of 800 K. An ATL Lasertechnik

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KrF excimer laser with a wavelength of 248 nm was used at a burst energy of 25 mJ and a frequency of 30 Hz. This setup yielded a growth rate of approximately 3 Å/min and cobalt ferrite layers with a thickness of 100 Å were prepared. The cobalt ferrite films did not show any contaminations in XPS measurements after growth.

The cobalt ferrite targets for PLD were synthesised by mixing of 3.000 g of cobalt(II) oxide and 6.402 g of iron(III) oxide, pressing of the milled powder and subsequent sintering at 1800 K for 6 h.

SQUID measurements were carried out with a Quantum Design MPMS-7. A diamagnetic contribution to all measured magnetic moments was assumed and was determined by fitting a straight line to the high field portion of the hysteresis loops. Subsequently the ferromagnetic signal was extracted by subtraction of the diamagnetic contribution. XPS measurements were carried out at room temperature with a VG Escalab 220i XL using Al K_{α} (1486.6 eV) radiation. For XRD measurements a Philips X'Pert Pro with a Cu K_{α} (8047.8 eV) source was used.

3. Results

After deposition of the cobalt ferrite films their quality was checked by means of photoemission spectroscopy (see supplementary data). The spectra showed no contaminations with carbon or other elements. Furthermore, the composition of the cobalt ferrite was checked by using the Co 2p, Fe 2p and O 1s levels for quantification. In these measurements a composition of $\text{Co}_1\text{Fe}_{1.9}\text{O}_4$ was found which is in good agreement with the stoichiometric composition of cobalt ferrite.

XRD measurements yielded a broadened (400)-peak, shown in Fig. 1, that corresponded to a lattice parameter $d=8.45$ Å which is an acceptable agreement with the value of $d=8.39$ Å found in literature for the bulk allowing for some epitaxial strain in the cobalt ferrite [10]. The broadening of the cobalt ferrite diffraction peak can be attributed to the very small layer thickness where diffraction only occurs at a small number of lattice planes [11]. The barium titanate substrate exhibited both (200)- and (002)-peaks corresponding to a and c domains, respectively. The small tetragonality of only 1.1% allows the cubic cobalt ferrite to grow on both structural domain types [12]. The impact of the tetragonality is even further limited as the growth is actually carried out at a substrate temperature of 800 K at which the barium titanate is in its cubic phase [8]. As the orientation of the barium titanate crystals is not controlled by applying an electrical field

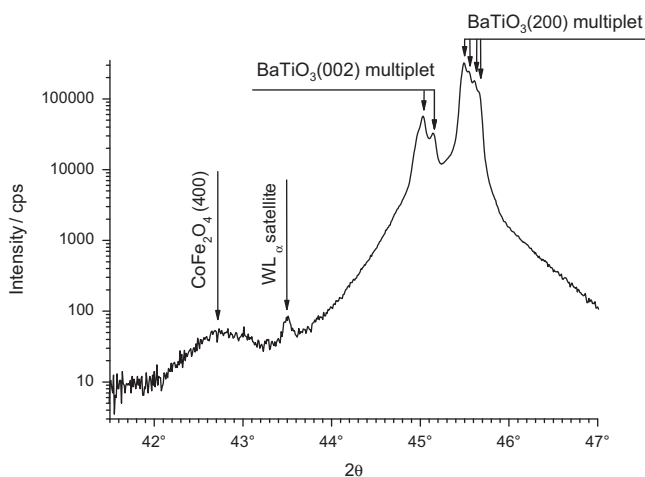


Fig. 1. X-ray diffractogram of a 100 Å cobalt ferrite film on barium titanate.

while cooling them through their phase transitions, their structural domains are distributed statistically within the macroscopic crystal [13,14]. The splitting of the two barium titanate peaks into multiplets can be attributed to two phenomena. On one hand there is the possibility of the occurrence of domain walls with finite size and lattice parameters within the domain walls that range between the values for a and c domains [15]. On the other hand the alternating a and c domains act as a sort of superstructure which in turn may result in satellite peaks [15].

Because of the crystal thickness of 1 mm, it was deemed unlikely that the shape of the crystal induces a preferential alignment of the structural domains. Therefore, we assumed that the barium titanate crystals behave magnetically isotropic in integral measurements as they average over all microscopic structural domains, albeit the microscopic structural domains individually might exhibit varying properties.

The hysteresis loops of the samples and of pure barium titanate at 100 K are shown in Fig. 2. The in-plane magnetisation of cobalt ferrite saturates at about 70 emu/g which corresponds well to the value of 75 emu/g found in literature [10]. It is to be noted that the cobalt ferrite film exhibits a large coercivity of 400 mT. On the other hand the out-of-plane magnetisation does not saturate at magnetic fields of 7 T and exhibits a much smaller coercivity of only 100 mT. This clearly indicates that the hard axis of the film is aligned perpendicular to the surface.

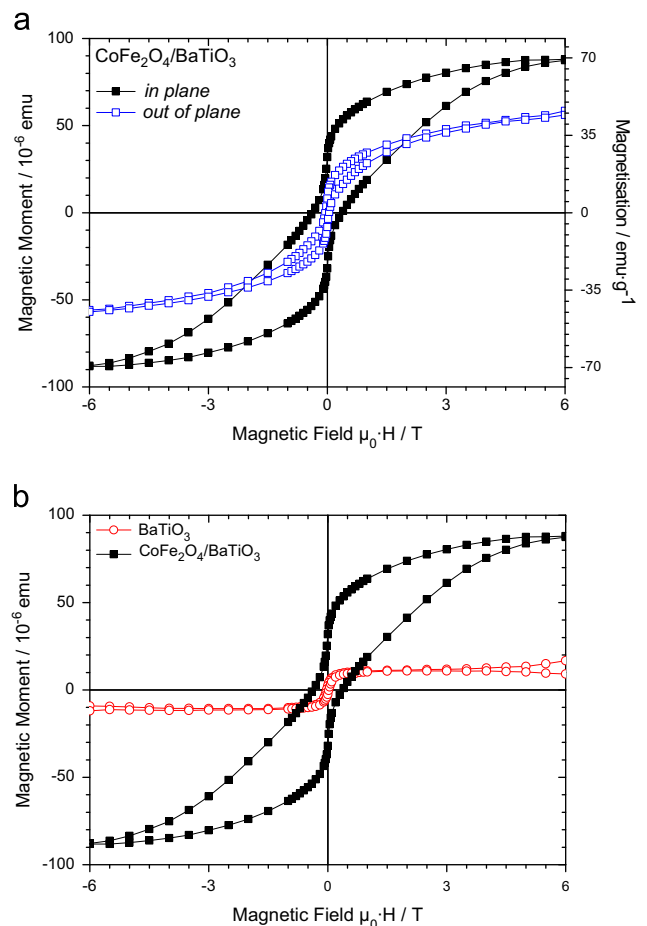


Fig. 2. Hysteresis loops of barium titanate before and after deposition of cobalt ferrite at a temperature of 100 K. In (a) the in-plane and out-of-plane hysteresis loops of a 100 Å cobalt ferrite film are compared and (b) sets the in-plane hysteresis loops of the bare barium titanate against the cobalt ferrite film on barium titanate.

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