Solution-deposited $BiFeO_3$ films: Photovoltaic effect & electro-optic response in dependence of doping & substrate stress

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Introduction

Bismuth ferrite ($BiFeO_3$, BFO) is special among perovskite-structured ferroelectrics, as it displays both ferroelectric and antiferromagnetic order. Both the high Curie temperature of more than 1000 K and the large spontaneous polarization in the order of $1\,C/m^2$ in BFO single crystals also make it attractive for basic scientific study and potential applications. Last, but not least, BFO displays fascinating photo-ferroelectric and optical properties such as a high birefringence and a pronounced anomalous photovoltaic effect. Photovoltaic conversion efficiencies several orders of magnitude higher than that for the bulk photovoltaic effect in other ferroelectrics were reported for epitaxial BFO films, spurring intensive research of BFO for photovoltaic applications [1]. For these investigations, BFO is usually in the form of epitaxial films created using vacuum deposition technologies such as pulsed laser deposition (PLD) or sputtering. A more cost-effective method to fabricate thicker BFO films is chemical solution deposition (CSD), e.g. by spin coating, typically resulting in polycrystalline films. One major drawback of these films is their comparably high conductivity, causing high leakage currents and making it difficult to achieve fully polarized samples.

Results

In this presentation, we will show how high-quality highly out-of-plane $(001)_{pc}$-textured polycrystalline BFO films can be prepared by spin-coating [2]. Leakage is drastically reduced by doping with Mn and Ti, resulting in films with properties that rival those of epitaxial films. The polarization hysteresis P(E) can be measured with frequencies down to kHz and below, and leakage-corrected remanent polarization is around $0.5\,C/m^2$, about 70 % of the maximum value possible for this orientation, based on single-crystal data. A variation of the substrate material allows to modify the mechanical stress in the film, ranging from 0.54 GPa tensile to 0.93 GPa compressive stress; the variation of the ferroelectric hysteresis at different stress states is minor.

Coplanar interdigitated electrodes (IDEs) are used to achieve electrical in-plane poling. Illumination with a wavelength of 455 nm generates a short-circuit photocurrent with a current density J_{sc}. Mn-doping causes an increase of the current density to the order of several tens of $\mu A/cm^2$; the sign of J_{sc} depends on the ferroelectric polarization direction. As can be seen in Fig. 1, the films display all characteristics of a bulk photovoltaic effect: J_{sc} and photoconductivity σ_{Ph} are linear with light intensity, but the open-circuit voltage V_{OC} is independent of intensity (Fig. 1(a) & (b)), V_{OC} scales with electrode distance (Fig. 1(c)), and most importantly, J_{sc} shows a sinusoidal dependence on light polarization angle, in agreement with the tensorial nature of the bulk photovoltaic effect. There is a strong dependence of the photovoltaic effect on in-plane mechanical stress, a phenomenon that is attributed to a piezo-photovoltaic effect [4].

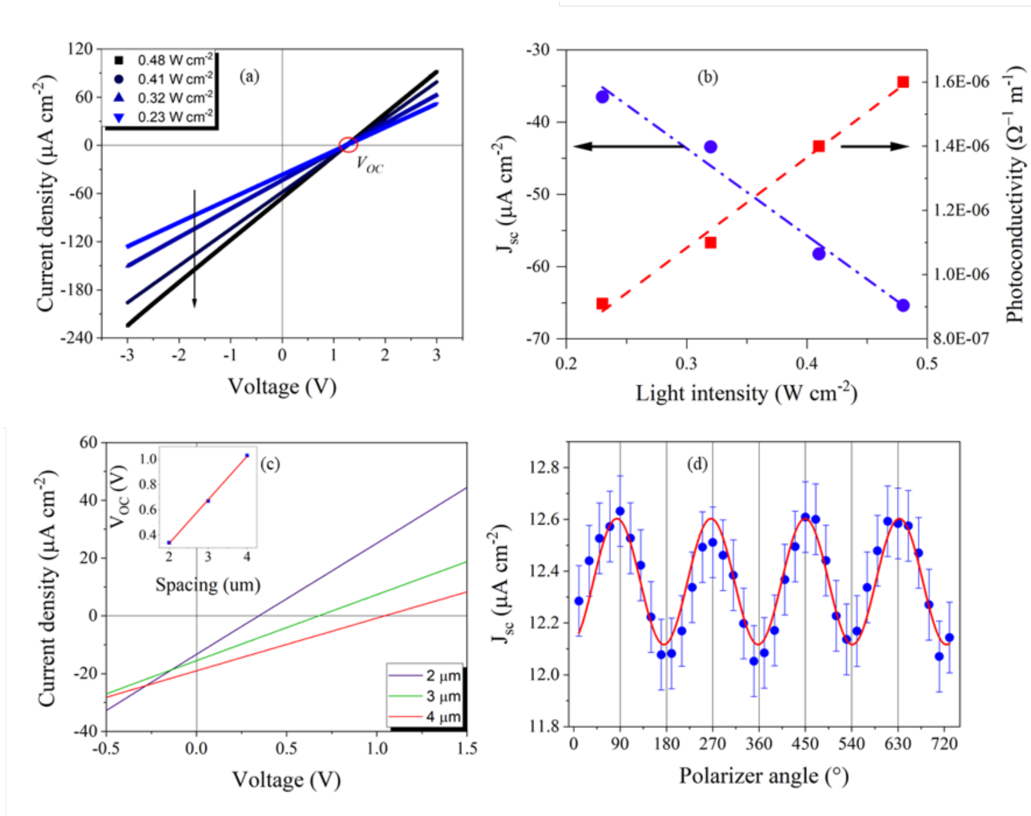

Figure 1: (a) Current-voltage curves of BFO at different light intensities. (b) Variation of J_{sc} and σ_{Ph} with light intensity (c) J-V curves of BFO with different electrode spacing (d) Variation of J_{sc} with light polarization direction at an intensity of $0.10\,\mathrm{W/cm^{-}2}$. 0° denotes the light polarization parallel to the poling direction. Reproduced from [3].

The Pockels effect of the films is quantified using a modified Teng-Man setup in transmission geometry. The values of the electrooptic tensor are notably lower than those of BFO single crystals and show little dependence on substrate stress or doping. Combined with the high photovoltaic effect and reasonable dark conductivity of the films, BFO seems a good candidate for transient photorefractive applications such as reconfigurable waveguides.

References

[1] G. Chen et al., Mater. Res. Bull. **110**, 39 (2019).
[2] Alfredo Blazquez Martínez et al., J. Eur. Ceram Soc. **41**, 6449 (2021).
[3] Alfredo Blazquez Martínez et al., Scripta Mater. **211**, 114498 (2022).
[4] S. Nadupalli et al., Sci. Adv. **5**, eaau9199 (2019).