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Electronic supplementary information

$La_xBa_{1-x}Ti_{1-x}Yb_xO_3$ (LBTYb) ceramics were prepared by the conventional mixed oxide route. The starting materials were high-purity grade $BaCO_3$, La_2O_3, TiO_2 and Yb_2O_3 powders (99.98%, +99.9%, +99.9%, and +99.99% purity, respectively, Aldrich Chemical Co., Milwaukee, WI). The powders were weighed in required molar ratios and intimately mixed by ball milling in acetone for 20 h, using yttria-stabilised zirconia balls. The dried slurries were passed through a 250 μm mesh and calcined in air at 1400ºC (exception for undoped $BaTiO_3$, x=0, which was calcined at 1200 ºC). Powder purity and crystallinity was determined by X-ray diffraction (XRD) using a high-resolution diffractometer ($CuK\alpha_1$, 1.54059 Å, Model Stoe StadiP, Stoe and Cie GmbH, Darmstadt, Germany) operated at 50 kV and 30 mV (step size of 0.02º and scan rate of 0.2º/min). The calcined powders were finely milled and uniaxially pressed (Specac, Kent, UK) into pellets under an applied pressure of ~40 MPa and then isostatically pressed (Model CIP 32330, Flow Autoclave System Inc., Columbus, OH, USA) at 200 MPa. The green compacts were fired in the temperature range 1400-1600ºC for 6 h in air, using a controlled heating cooling rate of 5ºC/min. The ceramics were mounted onto a rotational sample holder and their purity and crystallinity was also determined by XRD using a reflection diffractometer ($CuK\alpha_1$, 1.54059 Å, Model D500, Siemens, Germany). Ceramic microstructures of as-fired and thermal etched surfaces were examined using a Scanning Electron Microscope (SEM) (Model Jeol 6400, Jeol Ltd, Tokyo, Japan) operated at accelerating voltage of 20 kV and a working distance of 8 and 15 mm. Energy Dispersive Spectroscopic (EDS) analysis was carried out using a Oxford Link unit (Model eXL, Oxford Instruments, Oxon, UK) coupled to the microscope. For Transmission Electron Microscopy (TEM), ceramics were mounted on a glass slide using a thermosensitive resin and ground to approximately 20 μm thick after which a 3.05 mm Cu support ring with 800 μm circular hole was glued onto its surface using an epoxy resin. After removing from the glass slide the samples were thinned in an ion beam thinner (Model Duo Mill, Gatan, Pleasanton, CA) operating at an accelerating voltage of 6 kV and a combined gun current of 0.6 mA at an incidence angle of 15º until perforation. The samples were examined using a TEM (Model EM 430, Philips, Eindhoven, Netherlands) operated at an accelerating voltage of 300 kV. Energy Dispersive Spectroscopic (EDS) analysis

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was carried out using a Oxford Link unit (Model eXL, Oxford Instruments, Oxon, UK) coupled to the microscope.

Room temperature Raman spectra were obtained using a micro-Raman spectrometer (Model, Renishaw inVia, UK) with an Ar laser λ=514.5 nm to excite the sample and spectra recorded in backscattering geometry. Laser power of 4 mW was focused to ~2 μm within the grains of the ceramic samples.

Gold sputtered electrodes were applied to the major faces of the ceramic pellets. The dielectric properties were investigated over the frequency range 10^2 -10^6Hz using both an LCR bridge (Model HP4284, Hewlett-Packard, Palo Alto, CA) coupled to a closed-circuit He-cryocooler (model CC 1.5 Oxford Instruments, Oxfordshire, UK) for subambient measurements (~8–300 K). ε_r and tan δ at MW frequencies and room temperature was measured by the cavity method using the $TE_{01\delta}$ mode. Measurements were carried out with a vector network analyzer (R3767, Advantest Corporation, Tokyo, Japan). Magnetic measurements were performed on a ceramic with x=0.4 using a SQUID magnetometer (Quantum Design, model MPMS-XL). The temperature dependence of magnetization (*M*) was measured in the temperature range 2-300 K at an applied magnetic field (*H*) of 0.05 T upon heating samples under zero field-cooled conditions from 2 K.