

An *in-situ* Raman spectroscopic investigation in electric fatigue behaviors of PLZT ceramics

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Received 3 September 2015; received in revised form 1 October 2015; accepted 5 October 2015

Available online 19 October 2015

Abstract

A series of *in-situ* Raman spectra upon different polarization directions of incident and scattered lights (θ) and the corresponding hysteresis loops of unpoled lanthanum-doped lead zirconate titanate (PLZT) ceramics were simultaneously measured without and with applications of various electric fatigue cycle numbers (N). Accordingly, the relative Raman intensities in optical modes of E(2TO) over E(3TO+2LO)+B₁ ($I_{E(2TO)}/I_{E(3TO+2LO)+B_1}$), the maximum changes in $I_{E(2TO)}/I_{E(3TO+2LO)+B_1}$ at $\theta=0-360^\circ$ (ΔR_{max}) and the remnant polarization (P_r) were obtained for the as-received ($N=0$) and differently electric fatigued ($N=10^0, 10^4, 10^5, 10^6$ and $10^{6.2}$) PLZT specimens. The characteristic electric fatigue behaviors observed for the unpoled PLZT ceramics were quantitatively analyzed by the normalized remnant polarization denoted as P_r/NP_r , 10° and correlated to the average domain reorientation defined as $\Delta R_{max}/N\Delta R_{max}$, 10° . It was concluded that during the electric fatigue process, the randomly orientated domains in the unpoled PLZT specimen as-received might be reoriented preferentially along the direction of the applied electric field initially. The electric fatigue reduced the extent of average domain orientation and significantly deteriorated the ferroelectric properties.

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Keywords: Ferroelectric ceramics; *In-situ* Raman spectroscopy; Electric fatigue; Domain orientation

1. Introduction

The lead zirconate titanate (PZT) and lanthanum-doped lead zirconate titanate (PLZT) ceramics have been most widely used in piezoelectric devices for their superior piezoelectric properties over an extended range of operation temperatures. The fatigue and failure induced by alternative mechanical loads and/or electrical fields have significantly impeded commercial applications of ferroelectric ceramics [1]. Compared with the ferroelectric perovskite thin films, the information in fatigue behaviors of bulk ferroelectric ceramics are very limited due to the

complex nature of polycrystalline ferroelectrics and the lack of effective investigation methods. Although the effects of composition and thickness [2,3], temperature [3], frequency and magnitude of electric field [1,4,5], as well as type and duration of applied load [6] on fatigue behaviors of PZT/PLZT polycrystalline materials have been previously studied, the exact fatigue mechanism of bulk ferroelectrics ceramics remained unclear.

Domain switching has been recognized as the major cause for ferroelectric deterioration. Understanding the relationship between the microscopic domain and macroscopic ferroelectrics is a key for elucidating the fatigue mechanism. During the application of an electric field, the polarization states on the surfaces of polycrystalline ceramics vary. If the domain observation is not performed on the same grain, the obtaining results may not be comparable. Much effort has been made towards searching for an *in-situ*, micro-regional and quantitative analysis method for microscopic domain observation.

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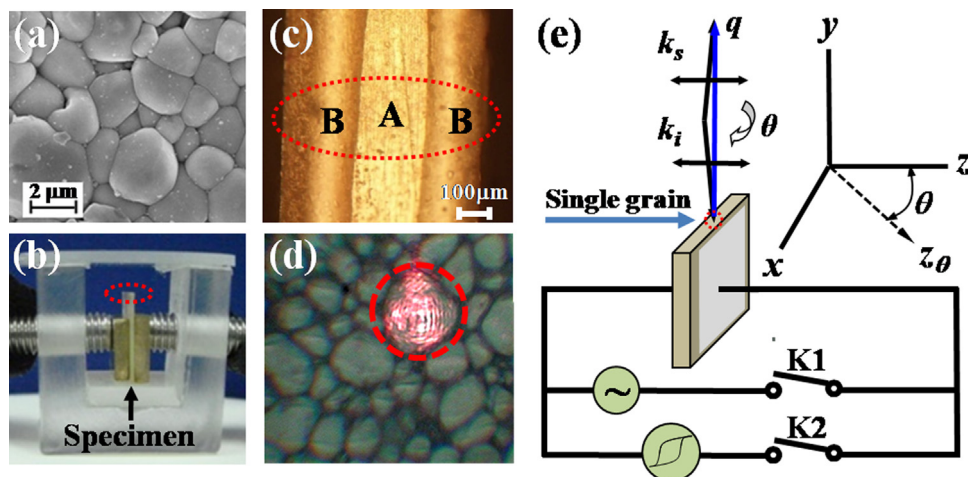


Fig. 1. (a) Micrograph showing typical grain sizes of PLZT ceramics, (b) photograph of a PLZT specimen electrode, (c) micrograph of the Raman observing surface, (d) photograph showing a probed grain with the focused laser beam, and (e) schematic drawing of the experimental setup with the backscattered geometry in the parallel $y(z_\theta, z_0)y$ configuration and the rotation angle θ for *in-situ* Raman spectroscopic and hysteresis loop measurements during an electric fatigue process.

Raman spectroscopy is a very attractive and non-destructive technique since optical modes are directly related to the spontaneous polarization of ferroelectrics, which reflects the physical essence of domain reorientation [7,8]. More importantly, the domain information on the same grain of polycrystalline materials may be obtained by *in-situ* Raman spectroscopy, thus, a comprehensive quantitative analysis may be performed and a comparable data may be collected. However, a particular experimental setup has to be designed to carry out the *in-situ* Raman spectroscopic measurements. In addition, compared with a single crystal material, an interpretation of Raman spectrum for a polycrystalline material has become extremely difficult as the Raman bands measured are always broadened and severely overlapped. Previously, a special 90° rotation device was established and employed to achieve the *in-situ* Raman observation of 90° domain switching for PLZT polycrystalline ceramics [9]. The number of 90° domain switching was characterized by evaluating the relative Raman intensity of E(2TO) and E(3TO+2LO)+B₁ modes ($I_{E(2TO)}/I_{E(3TO+2LO)+B_1}$) and further related to the electric fatigue behaviors of unpoled, in-plane and out-of-plane poled PLZT ceramics [9]. However, those correlations became ambiguous since the *in-situ* Raman spectroscopic and hysteresis loop measurements were not done with the same grain on the same specimen. Based on a series of *in-situ* Raman spectra measured for PLZT ceramics with various polarization directions of incident and scattered lights, represented by the rotation angles ranging $0\text{--}360^\circ$, the difference between the maximum and minimum $I_{E(2TO)}/I_{E(3TO+2LO)+B_1}$ values in the range of $0\text{--}360^\circ$ was defined as ΔR_{max} [10]. It was shown that the variation of ΔR_{max} with the applied electric field was nearly identical to that of the piezoelectric coefficient (d_{33}) of the samples polarized by different electric fields [10]. Since the magnitude of d_{33} indicates the degree of domain orientations, the ΔR_{max} was considered to signify the extent of domain reorientation induced by an electric field [10]. Unfortunately, the ferroelectric fatigue behavior has not been simultaneously studied.

In this work, a systematical investigation was conducted in an effort to collect the *in-situ* Raman spectra at room temperature on the as-received and different electrically fatigued PLZT specimens for various polarization directions of incident and scattered lights. Upon the completion of every Raman spectroscopic measurement, the corresponding hysteresis loops were also measured for the same specimens before and after different electric fatigue cycle numbers (N). The important parameters, $I_{E(2TO)}/I_{E(3TO+2LO)+B_1}$ and ΔR_{max} as described previously [9,10], as well as remnant polarization (P_r) were obtained for the same PLZT specimen under every differently electric fatigued condition ($N=10^0\text{--}10^{6,2}$), and then compared with those of as-received condition ($N=0$). The average domain orientation was evaluated and quantitatively related to the decayed ferroelectric properties represented by the normalized remnant polarization.

2. Experimental

2.1. Materials and electrode preparation

The unpoled PLZT ceramics were purchased from Jinghua Radio Component Factory of China, having an atomic ratio of Zr:Ti=52:48. The space group of $P4mm$ and the lattice parameters of $4.017 \times 4.017 \times 4.092 \text{ \AA}^3$ were determined by XRD Rietveld method with a typical average grain size of $2\text{--}4 \mu\text{m}$ as seen in Fig. 1(a). The specimens have a dimension of $0.5 \times 10 \times 12 \text{ mm}^3$ in a rectangular shape, with the silver layers covering the entire area of the two $10 \times 12 \text{ mm}^2$ surfaces served as an electrode for the subsequent electrical loading (Fig. 1(b)). Meanwhile, the Raman observing surface (circled in Fig. 1(b)) was covered by the insulating resin (B) except a small area (A) exposed for Raman observation (Fig. 1(c)). To avoid discharge between two electrodes upon the electric application, a margin of 1 mm along the three sides on both $10 \times 12 \text{ mm}^2$ surfaces away from the Raman observing surface of the specimen was created by nitric acid solution.

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