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Pure low-frequency flexural mode of $[011]_c$ poled relaxor-$PbTiO_3$ single crystals excited by k_{32} mode

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Rhombohedral phase relaxor-$PbTiO_3$ solid solution single crystals poled along $[011]_c$ exhibits superior lateral extensional piezoelectric response, which enables the excitation of a pure low frequency flexural mode with a bridge-type electrode configuration. For the ternary $0.24Pb(In_{1/2}Nb_{1/2})O_3$-$0.46Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.30PbTiO_3$ single crystal poled along $[011]_c$, the electromechanical coupling factor of the flexural mode reached as high as 0.66, and the resonance frequency of this mode can be easily made in kHz range, making it possible to fabricate very small size low frequency sensors and actuators. We have delineated theoretically the coupling between flexural mode and other modes and realized a strong pure flexure mode.
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Due to their excellent electromechanical properties, binary and ternary relaxor-$PbTiO_3$ solid solution ferroelectric single crystals, such as $(1\text{-}x)Pb(Mg_{1/3}Nb_{2/3})O_3$-$xPbTiO_3$ (PMN-PT) and $(1\text{-}x\text{-}y)Pb(In_{1/2}Nb_{1/2})O_3$-$yPb(Mg_{1/3}Nb_{2/3})O_3$-$xPbTiO_3$ (PIN-PMN-PT), have attracted considerable attention over the past decade.[1–8] Giant longitudinal piezoelectric coefficient $d_{33} > 2000$ pC/N and very high electromechanical coupling factor $k_{33} > 90\%$ could be obtained when these rhombohedral phase crystals were poled along $[001]_c$ of the pseudo-cubic direction. Recently, it was found that the shear piezoelectric coefficients d_{36} could excess 2000 pC/N for a 45°-z rotated relaxor-$PbTiO_3$ single crystal with engineered domain structures having *mm2* macroscopic symmetry,[3] which also has a very high electromechanical coupling factor of $k_{36} > 0.80$ for the related face shear mode.[9] Together with some unique characteristics, the face shear modes are very promising for applications in low frequency piezoelectric devices.

Another low-frequency mode of piezoelectric vibrators is the flexure mode. The flexural vibration of a bar (or a plate) can be generated when part of the bar (or plate) is stretching while other part of the bar (or plate) is compressing, or when two parts of the bar (or plate) have opposite shear strains.[10] Thus, this passive flexure mode can be excited through either the extensional or shear mode. The piezoelectric flexure elements, or bimorphs, were first introduced eight decades ago by Sawyer[11] and have been used in numerous applications, such as magnetoelectric coupling devices,[12] nanogenerators,[13] and piezoelectric composites.[14]

Due to the passive nature of the flexural vibration, the electromechanical coupling properties of this mode are determined by the active excitation mode. It was demonstrated recently that for some relaxor-$PbTiO_3$ crystals poled along $[011]_c$, the piezoelectric coefficient d_{32} could be as large as 1880 pC/N, and the electromechanical coupling factor k_{32} as high as 0.94.[15,16] Such excellent lateral extensional properties of $[011]_c$-poled relaxor-$PbTiO_3$ multidomain single crystals make it possible to excite a strong flexural mode, i.e., use the k_{32} mode as the active excitation mode to excite the passive flexural mode. In general, the flexural mode is often coupled to some low frequency modes, such as extensional and face shear modes, which lead to a significant performance degradation of the corresponding piezoelectric devices. Up to now, investigations on relaxor-$PbTiO_3$ single crystals have been mainly on active piezoelectric modes, such as longitudinal and transverse modes,[5,6] whereas no study has been carried out on flexural vibrations. This is because the flexural mode is often treated as spurious mode, so that people usually are trying hard to avoid exciting such modes.

In the present work, pure flexural vibration of ternary PIN-PMN-PT single crystal bar excited by the k_{32} mode has been investigated experimentally and analyzed theoretically. In addition, we have also studied the electromechanical coupling factor of flexural mode and its temperature dependence, and the mode coupling between the excited flexural mode and other active modes, such as extension and face shear excitations.

In our experiments, ternary 0.24PIN-0.46PMN-0.30PT single crystals grown by the modified Bridgman method were used. Each sample was cut and polished into a thin bar with rectangular cross-section. The orientations of the bar are $[011]_c$, $[0\bar{1}1]_c$, and $[100]_c$ along thickness, width, and length directions, respectively. These directions correspond to the z, x, and y directions of the orthorhombic coordinates. All orientations were checked by a Laue x-ray machine with an accuracy of ±0.5°. Gold electrodes were sputtered onto the $[011]_c$ faces of the samples, and the samples were poled under a 10 kV/cm field at room temperature. The crystals with engineered domains have orthorhombic *mm2* macroscopic symmetry after poling. In the orthorhombic coordinate system, the bars are k_{32}-type resonators. The dimensions of the resonators were specified by the IEEE standards on

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piezoelectricity. The resonance and antiresonance frequencies of the resonator were measured by HP4294A impedance-phase gain analyzer.

In order to activate pure flexural piezoelectric mode, the electrode configuration was designed as in Figure 1(a). The length of the four Au electrodes, A, B, C, and D, deposited on $[011]_c$ faces is the same as the crystal bar, but the width of the electrodes has been divided into two parts. Electrode A is connected to D, and electrode B is connected to C to form a bridge configuration. When an electric field is applied, the d_{32} piezoelectric effect makes one part of the bar to expand while the other part to contract, resulting in the bending of the bar, namely, excited a flexural vibration. The schematic flexural motion of a free standing bar is shown in Figure 1(b).

The wave equation for the flexural wave propagating along an isotropic solid bar has been derived by Rayleigh[17] under the assumption that the wavelength of the ultrasonic wave is much longer than the width and thickness of the bar, thus, the bending is small so that the shear strain and rotation inertia of the bar can be ignored. For the piezoelectric bar shown in Figure 1(a), if the applied electric field can be considered uniform, the governing equation for the flexural wave will be the same as that for isotropic bars because the direction of particle vibration is normal to the electric polarization.[18] For the piezoelectric k_{32} bars, the constitutive equation can be written as

$$S_2 = s_{22}^E T_2 + d_{32} E_3, \tag{1a}$$

$$D_3 = \varepsilon_{33}^T E_3 + d_{32} S_2. \tag{1b}$$

When the bending is small, we can assume that there are no shear strains and no twists in the bar. The central plane of the bar undergoes neither extension nor compression. Therefore, it is a neutral plane. The particle displacement is along $[0\bar{1}1]_c$ (x direction) and the longitudinal strain is given by[19]

$$S_2 = -\frac{y}{R}, \tag{2}$$

where y is the distance from the neutral axis and R is the curvature radius of the neutral plane. Then, based on the procedure by Rayleigh,[17] the amplitude of the formal wave solution for the particle displacement can be written as,

$$x(y) = A\cosh\gamma y + B\cos\gamma y + C\sinh\gamma y + D\sin\gamma y. \tag{3}$$

Here, $\gamma^4 = \left(\frac{\omega}{V\kappa}\right)^2$ with $V = \sqrt{\frac{Y}{\rho}} = \sqrt{\frac{1}{\rho s_{22}^E}}$ to be extensional wave velocity in the bar and $\omega = 2\pi f$ to be the angular frequency. For a free standing bar, the mechanical boundary conditions at the ends will give the resonance frequency as

$$f_n = \frac{2\alpha_n^2\kappa}{\pi l^2}\sqrt{\frac{Y}{\rho}} = \frac{2\alpha_n^2\kappa}{\pi l^2}\sqrt{\frac{1}{\rho s_{22}^E}}. \tag{4}$$

Here, l is the length of the bar, α_n is the nth root of the transcendental equation $\tan\alpha_n + (-1)^n\tanh\alpha_n = 0$ $(n = 2, 3\ldots)$. For our experiment, the radius of gyration is given by $\kappa = \frac{b}{\sqrt{12}}$, where b is width of the bar. Thus, the fundamental frequency ($n = 2$) is determined by

$$f_r = \frac{1.0279b}{l^2}\sqrt{\frac{1}{\rho s_{22}^E}}. \tag{5}$$

In our experiment, $l = 17.4$ mm, $b = 2.8$ mm, $s_{22}^E = 71.11$ m^2/Pn, and $\rho = 8100$ kg/m^3. From Eq. (5), the calculated resonance frequency is $f_r = 12.5$ kHz, lower than the resonance frequencies of any other modes in 0.24PIN-0.46PMN-0.30PT crystal bars with the same dimensions.

Figure 2 gives the measured resonance spectra of 0.24PIN-0.46PMN-0.30PT single crystal bars with the bridge-configured electrode (Figure 1(a)) at room temperature. The measured resonance frequencies is $f_r = 12.1$ kHz, which agrees well with the calculated result. The activation of a pure flexural vibration in this configuration is thus confirmed. Furthermore, the effective electromechanical coupling factor can be estimated by

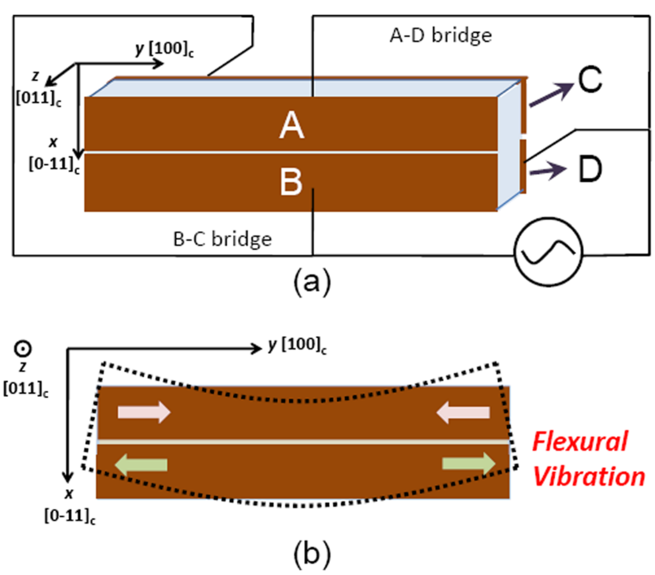

FIG. 1. (a) Bridge electrode configuration for activating the flexural mode; (b) Illustration of the flexural motion of a k_{32}–type bar.

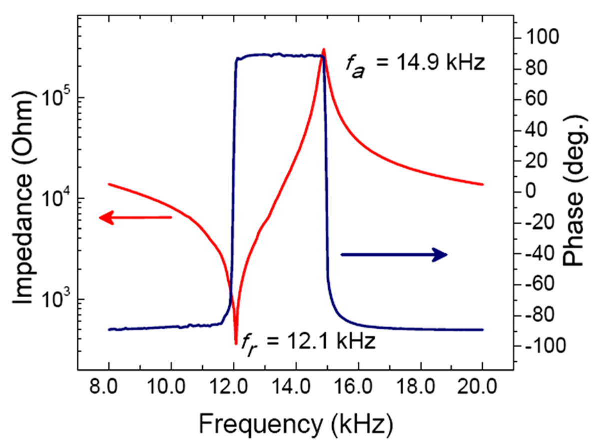

FIG. 2. Amplitude and phase spectra measured from a k_{32} extensional bar of 0.24PIN–0.46PMN-0.30PT crystal poled along $[011]_c$ with bridge electrode configuration.

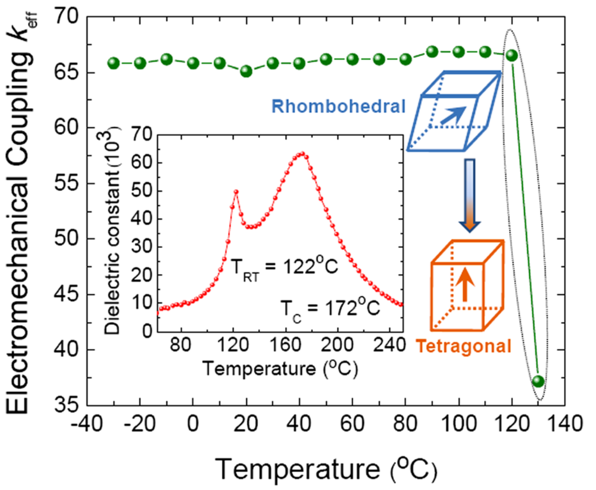

FIG. 3. Electromechanical coupling factor k_{eff} for flexural mode as a function of temperature. The small inset shows the dielectric constant $\varepsilon_{33}^T/\varepsilon_0$ as a function of temperature for 0.24PIN-0.46PMN-0.30PT single crystals measured along the poling direction $[011]_c$.

$$k_{eff} = \alpha\sqrt{\frac{f_a^2 - f_r^2}{f_a^2}}, \qquad (6)$$

where f_r and f_a are the measured resonance and antiresonance frequencies, respectively, α is a coefficient depending on the vibration mode and material constants. Now, the passive flexural mode is activated through the active mode of d_{32}, for which the coupling factor is related to the resonance and antiresonance frequencies: $\frac{k_{32}^2}{1-k_{32}^2} = \frac{\pi f_a}{2f_r}\tan\left(\frac{\pi(f_a-f_r)}{2f_r}\right)$. From this, α is estimated to be 1.127 for our sample. Based on this result, the effective electromechanical coupling factor of the flexural vibration of 0.24PIN-0.46PMN-0.30PT crystal bars is estimated to be around 0.66 at room temperature. Figure 3 presents the temperature dependence of the electromechanical coupling factor. It can be seen that k_{eff} is 0.65 at $-30\,°C$, increased only slightly to 0.66 up to 120 °C, at which the rhombohedral to tetragonal phase transition occurs. Above 120 °C, the coupling factor drastically decreased to 0.37. This temperature variation nature for the flexural electromechanical properties is quite similar to earlier reported results for the extensional and face shear modes.[2,9]

It is often observed that the resonance spectra of extensional bars or face shear plates are not so clean. Since flexural modes are generally the lowest frequency modes, we believe that the spurious modes appeared in most resonance spectra must be the flexural mode and/or its higher harmonics. For fully electroded extensional bars or face shear plates, the flexural vibration and its higher harmonics could be produced either by unbalanced stress caused by improper support or by unbalanced tractions on the surface of the bar or plate.[20] Figure 4 shows the resonance spectra measured for extensional bars and face shear square plates of 0.24PIN-0.46PMN -0.30PT crystals poled along $[011]_c$. One can see that mode couplings exist when the supporting points of the resonator are not proper. Clean resonance spectra could be obtained when the bars or plates are supported at their vibration nodes, or use conductive Cu base

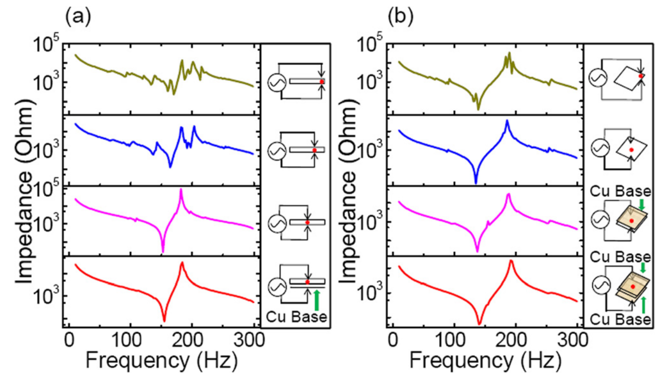

FIG. 4. Amplitude spectra measured under different support conditions for (a) extensional bars and (b) face shear square plates of 0.24PIN-0.46PMN-0.30PT crystals poled along $[011]_c$.

configuration to prevent the activation of the flexure modes.

In summary, by using a bridge electrode configuration, pure low frequency flexural vibration has been activated in a k_{32}-type bar of rhombohedral phase 0.24PIN-0.46PMN-0.30PT single crystal poled along $[011]_c$. The effective electromechanical coupling factor of the flexural mode reached ~0.66 at room temperature and remains stable up to the rhombohedral-tetragonal phase transition temperature. Superior to extensional and face shear modes, clean resonance can be easily established here since the resonance frequency of flexural mode is the lowest. More importantly, the driving electric field is parallel to the electric polarization of the crystals, so that the mode can endure high power driving. Therefore, our work opened up the door for both fundamental studies on the passive piezoelectric modes in relaxor-PT single crystals and for the applications of this material for low frequency ultrasonic transducers, piezoelectric motors, and acoustic sensors.

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