

# Domain Structure, Phase Transitions and Electric Properties of Novel $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3\text{-Bi}(\text{Zn}_{2/3}\text{Nb}_{1/3})\text{O}_3$ Piezo-/Ferroelectric Single Crystals

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$\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  (PMN-PT) single crystals have attracted considerable attention due to their high piezoelectric coefficient ( $d_{33} > 1500$  pC/N) and large electromechanical coupling factor ( $k_{33} > 90\%$ ) for the compositions near the MPB region, which entitles this system one of the most promising material for the next generation of electromechanical actuators and sensors for medical ultrasonic imaging and therapy, sonar transducers, and many other applications. However, the low temperature of the rhombohedral/(or monoclinic)-tetragonal phase transition (referred to as  $T_{R-T} \sim 60\text{-}95^\circ\text{C}$ ), low Curie temperature ( $T_C \sim 130^\circ\text{C}$ ) and low coercive field ( $E_C \sim 2.5$  kV/cm) make PMN-PT unsuitable for high temperature and high power applications. In this study, we incorporate  $\text{Bi}(\text{Zn}_{2/3}\text{Nb}_{1/3})\text{O}_3$  (BZN) into PMN-PT to form a new ternary PMN-PT-BZN solid solution in order to increase the  $T_C$  (and  $T_{R-T}$ ). Large-size PMN-PT-BZN single crystal were successfully grown by a flux method and their domain structure, phase transitions and electric properties were investigated. Different phase symmetries including rhombohedral, tetragonal and monoclinic phases were found to exist in these crystals, which is due to the composition segregation. The optical domain structure, dielectric, ferroelectric and piezoelectric properties of the crystals with various phases were studied. Especially an unusual phase transition sequence of rhombohedral-monoclinic-cubic was directly observed by polarized light microscopy (PLM). The dielectric measurements upon zero-field-heating reveal that the transition from the rhombohedral phase into a low symmetry phase (possibly monoclinic  $M_A$  phase) occurs at around  $50^\circ\text{C}$ , and the phase transition from monoclinic to cubic phase takes place at  $156^\circ\text{C}$ . An ultrahigh piezoelectric coefficient  $d_{33} > 2000$  pC/N was obtained from this crystal. The unusual phase transition sequence and the high piezoelectric response are explained by the polarization rotation mechanism. Based on these results, a temperature-composition phase diagram is established which delimits the various phases and the phase transition behavior. PMN-PT-BZN single crystals constitute a new piezocrystal system of ternary solid solution potentially useful for electromechanical transducer applications.

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