The lead-based relaxor ferroelectrics are chemically disordered perovskites that have remarkable structural, dynamical, and electromechanical properties. \( \text{PbMg}_{\frac{1}{3}} \text{Nb}_{\frac{2}{3}} \text{O}_3 \) (PMN) displays the broad, frequency-dependent peak in the dielectric response as a function of temperature that is characteristic of relaxors. Even though PMN possesses an unusually large dielectric susceptibility \( \varepsilon \sim 25000 \), it does not exhibit a well-defined structural transition, and thus no ferroelectricity, in zero-electric field.

An anomalous temperature- and wave vector-dependent damping produces a false, dispersion-like feature known as the “waterfall,” has been observed in PMN. At wave vectors away from the zone centre. To date, however, almost no interest had been taken in the zone boundary dynamics of the relaxors, since ferroelectricity is a \( \Gamma \)-point property. The structure of PMN and cubic-P Brillouin zone are shown in Fig 1. We decided to measure the dynamics along the \( M-T-R \) zone edge and began measurements on C5 and N5.

Our initial hypothesis was that classical tilt instabilities of the perovskites may couple indirectly to the ferroelectric activity, and a line of such modes is known to exist along \( M-T-R \). We found considerable low-frequency activity at the zone boundary, including columns of scattering that descend from the phonon at M and R from the C5 and N5 data. (Such columns have been rarely seen in insulators, although another example that was discovered at CRL was that of calcite by MT Dove and coworkers. A famous non-insulator example is beneath the dip in the dispersion of \( \beta \)-Ti and Zr, related to \( \omega \)-phase formation in these metals.) Clearly such modes are not normal modes, since a phonon can have only one frequency per wavevector. Later we extended these measurements on SPINS and DCS at NIST (Fig. 2), from which the effect can be more clearly seen in terms of a contour plot (Fig. 3).

The columns soften at 400 K, with a corresponding maximum in their intensity (Fig. 4), similar to the onset of the zone-centre diffuse scattering, indicating a coupling between the ferroelectric and the zone boundary activity. As the modes run along \( M-T-R \) and taking into account the observed splitting of the degeneracies at R, our initial idea was that these modes...
were the tilt modes, since they are commonly active in perovskites. However, for the zone in which the first measurements were made, the structure factors for the tilt modes are zero. Therefore, tilt modes cannot be responsible for this activity. From a study of the published dispersion curves of many perovskites, and from direct structure factor calculations of modes in many zones, we concluded that the M-T-R branch is a set of soft antiferroelectric modes $R_4^--T_1^-M_3^-$. $R_4^-$ involves Pb and O motion (Fig. 5), whereas $M_3^-$ consists only of Pb motions. This result could be explained in terms of coupling of antiferromagnetic modes to a primary ferroelectric order parameter. However, from comparison to other systems in which columns are known to exist (e.g. calcite), the antiferroelectric modes are more likely to be order parameters that compete with the ferroelectric ordering.

---

**Fig 4.** CS data showing the intensity of the M-point column as a function of temperature, showing a maximum near 400 K.

**Fig 5.** Eigenvectors of the low-frequency mode at $R$.

---

**References**