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Multiferroic Pb(Fe_{1/2}Nb_{1/2})O₃ Single Crystals: A Raman scattering study

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Among complex perovskite-type structure multiferroics $[A(B'B'')O_3]$, lead iron niobate Pb(Fe_{1\2}Nb_{1\2})O₃ (PFN) discovered by Smolenskii *et al.*¹ is currently of interest because of its high dielectric constant diffuse phase transition² and magnetoelectric effect^{3,4}. PFN presents ferroelectric transition below $T_{\rm C}$ ~350-395 K and antiferromagnetic transition below $T_{\rm N}$ ~145 K¹⁻⁴.

In PFN, the Pb2+ cations with a lone electron pair in A site and the relative displacements of oxygen and Fe³⁺/Nb⁵⁺ ions in the octahedral B sites favor the electric ordering whereas the Fe³⁺ cations with partially filled d orbitals contribute to the magnetic ordering. Because of its complex structure, still exists a disagreement about the crystalline structure of PFN: monoclinic (space group Cm), rhombohedral (space group R3m), and cubic (space group Pm3m) structure symmetry at room temperature were reported^{3,5,6}.

In this work, we report temperature-dependent Raman spectra (10-500 K) in the frequency range of 150-1250 cm⁻¹ of cubic PFN single crystals, complemented with magnetization measurements. We want to provide microscopic information in this material.

Over the range from 150 to 950 cm⁻¹, very broad and overlapping peaks (first-order character) were observed in the overall studied temperature range possibly due to local distortions which are characteristic in this kind of Pb-based complex perovskite. Also, a prominent high-frequency peak at ~1130 cm⁻¹, which we assign as a two-phonon peak, was observed (see Fig. 1).

Temperature dependence of the Raman spectra showed an anomaly in a characteristic temperature $T^* \sim 330$ K. Here, drastic changes in the Raman scattering intensity and spectral shape were observed with temperature variations, such as the appearance of new first- and second- order peaks below T^* . The temperature behavior of the two-phonon peak shows a strong electron-phonon interaction in PFN. Also, since it was reported weak magnetic ordering at and even above room temperature in

this material, the anomaly at $T^* \sim 330$ K by Raman scattering could be possibly suggesting an interplay between magnetic e ferroelectric orders.



Figure 1: Raman spectra as a function of temperature in Pb(Fe_{1/2}Nb_{1/2})O₃.

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