

Supplementary Information

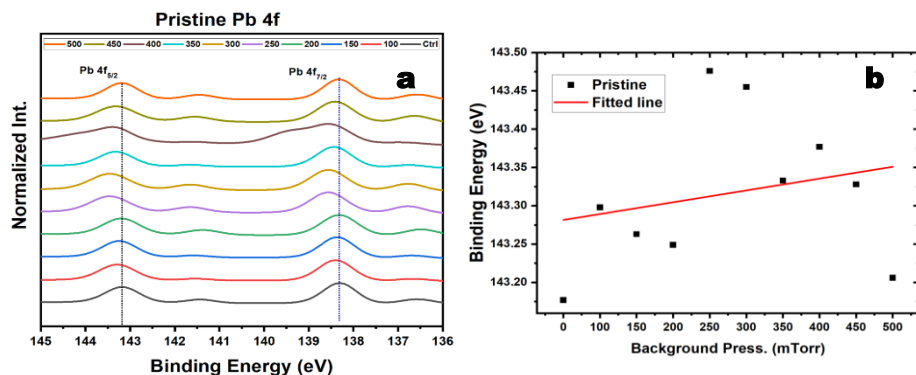


Figure 1. (a) Normalized Pb 4f spectra for pristine films deposited at different background pressures. The Pb^{2+} spectra show shifts to higher binding energies which is obvious at elevated background pressure. This is suggestive of changes in the chemical composition of the films due to increased interaction of gas molecules with the perovskite surface. (b) Scatter plot of change in binding energy vs background pressure. **0 mTorr and Ctrl represent the active vacuum condition around 0.01 mTorr.**

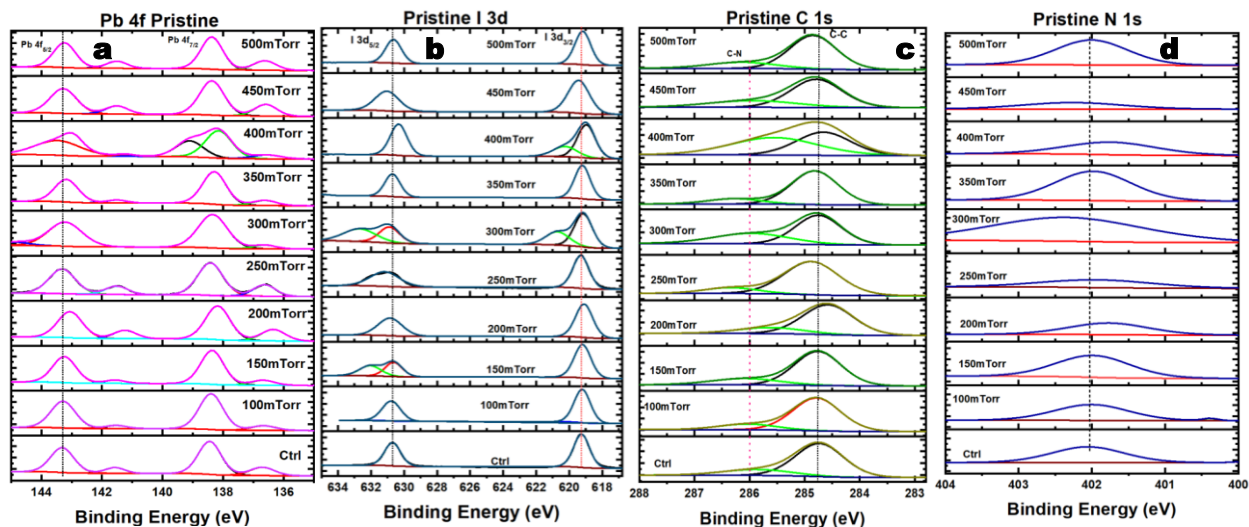


Figure 2. The high-resolution core level XPS spectra of (a) Pb 4f, (b) I 3d, (c) C 1s, and (d) N 1s for films deposited under different background pressures. Peak splitting is observed at 400 mTorr for Pb 4f and at 150, 300, and 400 mTorr for I 3d. This indicates that background pressure induces multiple chemical states resulting in various coordination environments of the films. **Ctrl represents the active vacuum around 0.01 mTorr.**

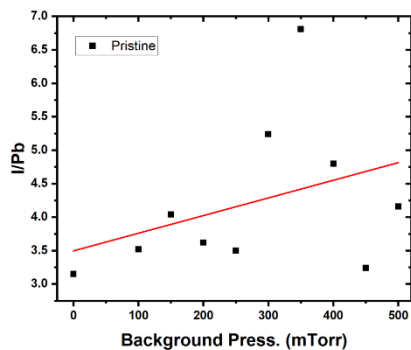


Figure 3. Scatter plot of I/Pb concentration determined from XPS as a function of background pressure. Excess I (ratio > 4) is observed at high pressures, resulting in non-stoichiometric conditions, while the control condition (ratio < 4) indicates vacancies.