Supplementary Information

Video S1: A 400 μm RM734 droplet on a bare glass substrate upon cooling from 130 to 80°C. In the first part of the video, the droplet undergoes a sort of "oscillatory" deformation toward the LN crystal and a weak electromechanical instability that leads to the ejection of two small fluid jets from the droplet side in front of the LN slab. When continuing to cool, a larger deformation occurs, which persists and eventually results in a violent instability with the ejection of large fluid jets from the whole droplet rim.

Video S2: A 308 μm RM734 droplet on a fluorolink-coated glass substrate upon cooling from 135 to 90°C. The N-N_F phase transition is visible in the first part of the video. Upon entering the ferroelectric phase, the droplet undergoes several instability events characterized by the emission of very thin jets that disrupt by forming a large number of small secondary droplets around the original one, and by the ejection of a large jet directed to the LN crystal. This latter produces a deformation of the droplet in the direction of LN and retracts upon leaving a part of the fluid material in contact with the LN border. The electrostatic interaction between the original droplet and this ferroelectric fluid mass gives rise to a "pulsing" instability consisting in the formation of thin jets and additional tiny droplets, and to the further ejection of a large jet toward the LN edge. By the end of the video, the fragmentation of the whole original droplet in secondary small drops is observed, together with several thin jets coming from the thin LC layer formed between the glass and LN during the previous instability events. Noteworthy, the fluid motion inside the droplet in the direction of LN is visible before the beginning of the instability.