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Time-Reversal Symmetry Breaking in Superconductors through Loop Josephson-Current Order

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Recent muon-spin relaxation experiments have found broken time-reversal symmetry (TRS) in a number of superconductors which from other points of view (such as specific heat, penetration depth, sensitivity to impurities etc.) appear to be conventional. We propose a novel superconducting groundstate where Josephson currents flow spontaneously between distinct, but symmetry-related, sites within a unit cell. Such Loop Josephson Current (LJC) state break TRS without the need for triplet, inter-site, or inter-orbital pairing i.e. they are compatible with a conventional BCS-type pairing mechanism. The Josephson currents result from a non-trivial phase difference between the on-site pairing potentials on different sites appearing spontaneously at the superconducting critical temperature. We show explicitly how such instability emerges in the Ginzburg-Landau theory of a simple toy model. We estimate the size of the resulting spontaneous magnetization and find it to be consistent with many existing experiments. We discuss the crystal symmetry requirements and apply our theory to the recently discovered family of TRS-breaking, but otherwise seemingly conventional, family of superconductors Re_6X ($\text{X}=\text{Zr}, \text{Hf}, \text{Ti}$), showing the possibility of a LJC instability