You can be made and broken in cycles, Mr. C-C Bond! The cyclobutane ring can easily be made by solid-state [2+2] cycloaddition reaction. But can this be cleaved? Is it possible to make and break the C-C bond reversible? Can this be achieved in a single-crystal to single-crystal manner? Will this be accompanied by change in properties like photoluminescence? If so the applications for these types of systems are eminent in molecular switching and data storage. The high thermal stability of an alkali metal-organic framework compound has successfully used for this purpose. It has been shown how the crystal packing controls the retention of single-crystal nature during cyclobutane formation and cleavage. The polymorph formed at high temperature retains its single-crystal (SC) nature during reversible photochemical formation and thermal cleavage in a SCSC manner, whereas the polymorph obtained at room temperature does not.