Topological Analysis of Charge Densities in Polymorphs of 3-acetylcoumarin

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Detailed investigation of the charge density distribution in concomitant polymorphs of 3-acetylcoumarin [1] in terms of experimental and theoretical densities show significant differences in the intermolecular features when analyzed based on the topological properties via the quantum theory of atoms in molecules [2]. The two forms, triclinic (form **A)** and monoclinic (form **B**) crystallize in centrosymmetric space group, form **A** in $P\bar{1}$ (Z'=2) and form **B** in $P2_1/n$ (Z'=1) and pack in the crystal lattice via weak C-H…O and C-H…π interactions. The electron density maps in two forms demonstrate the differences in the nature of the charge density distribution in general. The net charges derived based on the population analysis via multipole refinement and also the charges evaluated via integration over the atomic basins and the molecular dipole moments show significant differences between the two forms. The lattice energies evaluated at the HF and DFT (B3LYP) methods with 6-31G** basis set for the two forms clearly suggest that form **A** is thermodynamically stable compared to form **B**. Mapping of electrostatic potential over the molecular surface showing dominant variations in the electronegative region bring out the differences between the two forms.

[1] Munshi P., Venugopala K. N., Jayashree B. S., Guru Row T. N., *Cryst. Growth Des.*, 2004, **4(6)**, 1105. [2] BaderR. F. W., *Atoms in Molecules: A Quantum Theory,* Oxford University Press, Oxford, UK, 1990.

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