Investigation of Phase Transitions in the Fullerites C_{60} and C_{70} by Adiabatic Scanning Calorimetry

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Adiabatic scanning calorimetry (ASC) is a very unique method to measure, at extremely low heating (cooling) rates the enthalpy and heat capacity shape of a phase transition with very high precision [1].

We mainly report on an investigation of very pure sublimed C_{60} samples of different sources. Early experiments revealed that at high temperatures C_{60} crystals have a fcc structure (space group $Fm\bar{3}m$). Although the molecules are orientationally disordered, they are not completely free, but undergo hindered rotations due to the non-zero crystal field of the neighbouring molecules. Neighbouring molecules prefer orientations in which electron poor regions (pentagons) face electron rich locations (hexagons) on the neighbours. At 260 K a phase transition occurs to another cubic structure: the Pa3-phase is formed. Below 260 K orientational order gradually sets in. Our data show [2] that the transition temperature, the full width at half maximum and the height of the C_p peak are rate dependent even at heating (cooling) rates of the order of about 100 mK/h, i.e. 300 to 1500 times slower than used in standard commercial differential scanning calorimeters (DSC) and 15 to 150 times slower than recently reported in modulated differential scanning calorimetric (MDSC) experiments. This indicates that even at such low rates the material is not fully in a thermodynamic equilibrium state and behaves as a system with a very long internal relaxation time (of the order of 10 h) in completing the first-order phase transition. We will compare the C_{60}-results with adiabatic scanning calorimetric data we recently obtained on a very pure C_{70} sample, showing a phase transformation from a cubic structure at high temperatures to a rhombohedral structure at about 340 K.

[1] J. Thoen, *Phys. Rev*. A **37**, 1754 (1988).
[2] G. Pitsi, J. Caerels and *J. Thoen, Phys. Rev*. B **55** (1997) in press.