“Magnetic carbon nanocluster growth in a plume formed by MHz-pulse-rate laser ablation”

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Abstract: Material parameters of a solid experience a dramatic change when cluster size becomes less than a certain critical value. Hence, one can control material properties through control over cluster size. Previously we have reported a unique nanostructured carbon nanofoam, which has strong paramagnetic properties. Carbon nanoclusters produced by high-repetition-rate laser ablation of graphite and glassy carbon, which is typical diamagnetic material, exhibits para- and ferromagnetic behavior up to 90 K. We aim to control the properties of carbon nanofoam by changing the size of carbon nanoclusters, which are the “building blocks” for this material.

Conventionally, using low-repetition rate ns laser pulses, clusters are formed through the interaction of a laser-ablated plume in a noble gas through diffusion-limited aggregation. The gas fill serves as confinement for the ablated atomic plume reducing its diffusion velocity and therefore retaining atoms at a temperature and density appropriate for atom-to-atom sticky collisions, and cluster formation.

We present the results on carbon cluster formation in a laser plume formed by high repetition rate, in the range 0.15 MHz – 28 MHz, 12-ps laser ablation of graphite and glassy carbon targets. We demonstrate experimentally and describe theoretically that the carbon nanoclusters can be created by single picosecond laser pulses in the conditions when the time gap between the pulses is in the sub-microsecond time domain. We demonstrate that the time for the plume’s adiabatic expansion in vacuum appears to be sufficient for the many successive collisions to occur that result in expansion-limited aggregation of nanoclusters.

In conclusion we present magnetic susceptibility data shows large differences between zero field-cooled and field-cooled experiments that are consistent with formation of a spin glass-like state with unusually high freezing temperature. Magnetic inhomogeneity is supported by detailed EPR studies, where we recognized three different types of center with significantly different relaxation times, from very long one of the order of 1 ms, down to 100 ns. The measured high g-factors (> 2.003) of two of the centre types are typical of amorphous carbon systems with significant sp³ character, i.e. strongly non-planar parts of a carbon sheet.