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Cluster Growth in High-Power Picosecond MHz-rate Laser Ablation

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We have reported that ablation of graphite and glassy carbon targets by MHz repetition rate trains of picosecond pulses in ambient atmosphere 1-100 Torr results in formation of carbon clusters with unusual magnetic properties.[1,2] The carbon vapour temperature in the formation zone of the laser plume is in the range 1-10 eV, so the formation process takes place in partly ionized carbon plasma. The new conditions of the cluster formation by high repetition rate laser are now strongly dependent on the repetition rate, scanning rate and Argon flow velocity in the chamber. The cluster formation time and the size of the clusters are defined by the combination of these parameters.

The measured mass magnetization is positive, and the curves show paramagnetic-like behavior at low temperature up to ~92 K. Nevertheless, we observe a slight hysteresis with a well-defined coercive force at low temperatures as expected for a ferromagnet. Saturated magnetization $M(H)$ of 0.42 emu/g is equivalent to $9.0\times10^{-4}\mu_B$ per carbon atom. In a ferromagnetic system ($1\mu_B$ per spin), this value would correspond to about 1 unpaired spin per 1000 carbon atoms. Low-temperature ESR measurements give the concentration of unpaired spins as 1.8×10^{20}/g (3.6 unpaired spins per 1000 carbon atoms), in order-of-magnitude agreement, but somewhat higher than predicted for a completely ferromagnetic system. We suggest the presence of ferromagnetic domains in a predominant paramagnetic matrix. The analysis of impurities contribution to the magnetic moment suggests that the observed behaviour is an intrinsic property of the carbon nanoclusters.

In this paper we investigate the conditions in the laser plume via optical spectroscopy and time-resolved imaging. Such measurements enable us to gain an understanding of the fundamental processes determining the growth of nano-materials in non-equilibrium conditions such as those existing in our laser produced gaseous plume. Through understanding of the growth mechanisms we will gain control over the precise structure formation, and hence the ability to create material with tailored properties. We will report preliminary results on the nanocluster growth process through analysis of the diffusion of atoms and clusters in the plume; the temperature distribution in the plume; the growth rate of the nanoclusters; and cluster size distribution in various laser and background gas conditions.

1. A. V. Rode, E. G. Gamaly, A. G. Christy, J. D. Fitz Gerald, S. T. Hyde, R. G. Elliman, B. Luther-Davies, A. I. Veinger, J. Androulakis, J. Giapintzakis, *Unconventional magnetism in all-carbon nanofoam*, Phys. Rev. B, **70**, 054407 (2004).
2. A. V. Rode, E. G. Gamaly, A. G. Christy, J. D. Fitz Gerald, S. T. Hyde, R. G. Elliman, B. Luther-Davies, A. I. Veinger, J. Androulakis, J. Giapintzakis, *Strong paramagnetism and possible ferromagnetism in pure carbon nanofoam produced by laser ablation*, Journal of Magnetism and Magnetic Materials, **290-291**, 298-301 (2005).

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