Edge states versus in-plane defects in graphite magnetism

T. Makarova[1], V. Zagainova[1], A. Okotrub[2], L. Bulusheva[2], G. Chekhova[2], D. Pinakov[2], N. Yudanov[2], A. Shames[3]

[1] *Umea University, Sweden*
[2] *Nikolaev Institute of Inorganic Chemistry SBRAS, Russia*
[3] *Ben-Gurion University of the Negev, Israel*

Inclusion compound C2F with the molecules intercalated between layers of graphite fluoride matrix represent a system of loosely bound bilayer graphenes and serve as an excellent model for studying low-dimensional magnetism. We have experimentally studied correlation of the magnetic properties of the samples with the edge structure and the in-plane structure. Fluorination starts from the edges CF termination which causes strong paramagnetism, and the samples obey the Curie-Weiss behaviour. Further fluorination forms CF2 edges which drastically reduce localized spin concentration. At these stages the samples exhibit only low-temperature (< 3K) short-range antiferromagnetic ordering. This is consistent with the predictions that magnetism originates from the localized edge states that give rise to a high density of states at the Fermi level rendering spin-polarization instability. The 1D system is sensitive to thermal disorder destroying the spin arrangement, and magnetism appears only at low T. When fluorine starts to attack the basal plane, it results in strongly coupled localized magnetic moments. The mechanism of fluorination assumes the random attachment of primary fluorine atom to graphite network and further random direction of chain formation. Fluorine fills the basal planes forming both zigzag and armchair chains with the stoichiometry CF-CF-CF-CF alternating with the chains of non-fluorinated C-C-C-C. The areas of formation of C2F structure have limited size and form domains consisting of the C2F bands ordered at different angles. When the formation of chain structures inside the basal planes starts, a kink appears on susceptibility curves at about 150 K. When the wet process is finished, the fluorine atoms continue reorganizing on the basal plane: isolated fluorine atoms diffuse rapidly along a path above C-C bonds till the structure reaches the energetically preferable state. The chains are self-organizing, the metastable lone spins disappear and finally the temperature dependences of magnetic properties does not have anything in common with paramagnetism: susceptibility decays exponentially at low temperatures, which is a characteristic feature of a spin-gap system. Simultaneously a ferrimagnetic contribution to susceptibility is developed due to uncomplete cancellation of magnetic moments; the latter is seen from the M(H) curves. Thus, we observe the formation of bulk magnetic carbon in a self-assembly process. Intrinsic nature of novel carbon magnetism results in its sensitivity to the thermal treatment.

Heating up to 400 K destroys antiferromagnetically ordered chains and simultaneously destroys bulk magnetic order. On ageing, room-temperature ferromagnetism spontaneously restores; carbon magnetism is switched ON/OFF. ESR studies and ab initio calculations provide the evidence that magnetic units are the areas of delocalized ?-system between the fluorinated chains.