Final Report (DE-FG02-00ER45817)

Using the DOE support we have shown the following:

- We performed a systematic microscopic investigation of two completely dissimilar materials (namely, ZnO and rhombohedral-C_{60} polymers) exhibiting ferromagnetism in the presence of defects, and showed that this new phenomena has a common origin and the mechanism responsible can be used as a powerful tool for inducing and tailoring magnetic features in systems which are not magnetic otherwise. Based on our findings we proposed a general recipe for developing ferromagnetism in new materials of great technological interest. Our results support the role of complimentary pairs of defects in inducing magnetism in otherwise non-magnetic materials belonging to two widely differing classes with no apparent correlation between them. In both classes, ferromagnetism is found to be enhanced when the two kinds of defects form structures (pathways) of alternating effective donor and acceptor crystal sites leading to the development of electron charge and spin density like waves.

- Using ab initio density functional theory calculations we predicted the existence of a new class of carbon cages formed via hybrid connection between planar graphene sheets and carbon nanotubes. The resulting novel structure has the appearance of "nano-drum" and offers the exciting prospect of integrating useful device properties of both graphene as well as the nanotube into a single unit with tunable electronic properties. Creation of a hexagonal hole in the graphene portion of this structure results in significant magnetic moments for the edge atoms. The structure appears to be capable of sustaining ferrimagnetic state with the assistance of topological defects.
  The charge and spin distributions obtained in our calculations for the nano-drums are in striking contrast to those in planar graphene nanoribbons with a central hole. In this case, the central hole appears as the complimentary defect to those of the ribbon edges. Similar situation is found in case of the nano-drum in which the complimentary to the hole defects appear to be the pentagons along the curved surface of the drum. Charge oscillations found in the nano-drum are minimized in the nanoribbons. But more importantly, the hole edge atoms in the nano-drums retain significant magnetic moments; almost twice those of the corresponding ones in hydrogenated graphene nanoribbons (H-GNRs). These results suggest that the topological defects in the nano-drums may act like blocks to keep magnetic moments from "leaking" out from the hole defects. This may have significant implications for the the use of nano-drums in magnetic storage technology where the ratio, magnetic-moment/weight, is of paramount importance in any futuristic device applications.

- One of the basic problems of the DFT/LSDA+U theory is the efficient evaluation of the U-term. With this in mind we proposed an alternative approach for its calculation which is based on the knowledge of the Hartree-Fock wave functions of the system under consideration. As a result, the proposed approach is closer to the basic definition of the DFT/LSDA+U scheme and its hybrid-DFT nature. According to our approach, the U value is obtained in a consistent and ab-initio way using the self-consistently calculated wave functions of the given system at the level of the HF approximation. Our method is applicable for systems which include more than one type of elements with localized d-orbitals. The method has been applied the case of the doped Zn(Co)O systems successfully.

- Currently, theories based on conventional superexchange or double-exchange interactions cannot explain long range magnetic order at concentrations below percolation threshold in dilute magnetic semiconductors. On the other hand, the codoping induced magnetism, which can justify magnetic interactions below percolation threshold, has eluded explanation. With this in mind, we proposed that defect-induced magnetism in codoped non-magnetic materials can be viewed within a molecular generalization of the atomic double-exchange and superexchange interactions applied to an arbitrary bipartite lattice hosted by (or embedded in) defect-free non-magnetic materials. In this view, the crucial factor for the development of magnetism appears to be the defect-complementarity of the

codopants. We demonstrated this by taking ZnO and GaN (the most widely studied ferromagnetic oxide and nitride, respectively) as host materials and performing theoretical calculations using ab initio methods after codoping them with transition metal impurities for a variety of configurations. Our results indicate that the magnetic coupling among the induced and/or doped magnetic moments takes the form of an interaction among spin polarized molecular units which is facilitated by the formation of the hosted bipartite codopant structures [1]. The universality of the proposed mechanism is further supported by earlier results referring to the rhombohedral C_{60}-based polymers[2,3].

Our work over the last three years has led to several publications in quality journals[1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. The expertise gained in the prior investigations will provide valuable insights, contributing to the success of the present project.

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