Structural, electronic and spin properties of two NV centers in nanodiamond : quantum chemical simulation

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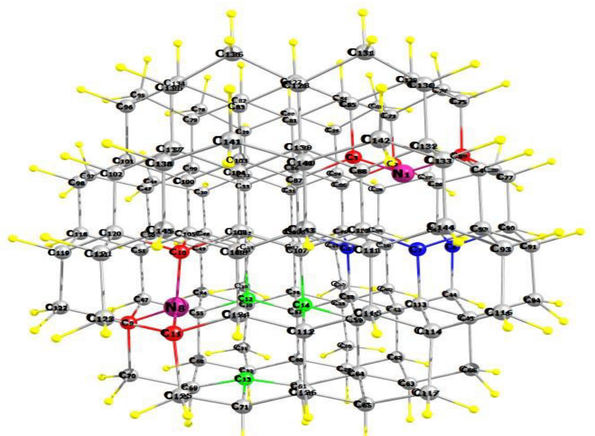
Motivation: A quantum antenna consisting of two two-level emitters can be implemented using a pair of color centers in diamond. The most famous and studied among them is the nitrogen-vacancy coloring center (NV center), which has a unique set of interrelated photophysical and spin properties, which are used for a variety of applications of NV centers. In this regard, it is relevant to perform direct computer modeling by methods of quantum chemistry of the system of two NV centers located not far from each other in the diamond cluster..

Our approach: Here we present the results of computer quantum chemistry simulation of the structure, electronic and spin properties of nanostructure , which consist of a nanodiamond (ND) containing a two nitrogen-vacancy centers (2NV). DFT and TD DFT calculations were performed using the Gaussian16 and ORCA 5.0.3 software packages.
As the object of study, a carbon cluster C_{143}[2NV]-H_{102} was chosen, containing two negatively charged NV centers oriented in different ways. The dangling bonds on the surface of the cluster were passivated by hydrogen atoms. The multiplicity for this cluster was taken to be 5 (quintet state, S=2), and the charge was twice negative. After the introduction of two replacement nitrogen atoms and the creation of a vacancy near each of them, the spatial structure of the resulting cluster was relaxed.

Fig. 1 - Schematic representation of a hydrogen-passivated diamond cluster C143[2NV]-H102 . Carbon-substituting nitrogen atoms are shown in yellow, nitrogen atoms in purple, and those closest to the vacancies of the first N1V1 and second N2V2 centers are shown in blue and green, respectively.



(Fig.1)

In a number of publications, the entanglement of the electron spins of two NV centers in the basic electronic state has been realized by coherent manipulation of the states of two adjacent NV centers interconnected by the interaction of magnetic dipoles. Therefore, within the framework of the density functional theory (DFT), the main value that determines spin-spin interactions in such a system was calculated, namely the distribution of spin density on a diamond nanocrystal containing two NV centers. (Fig.2)

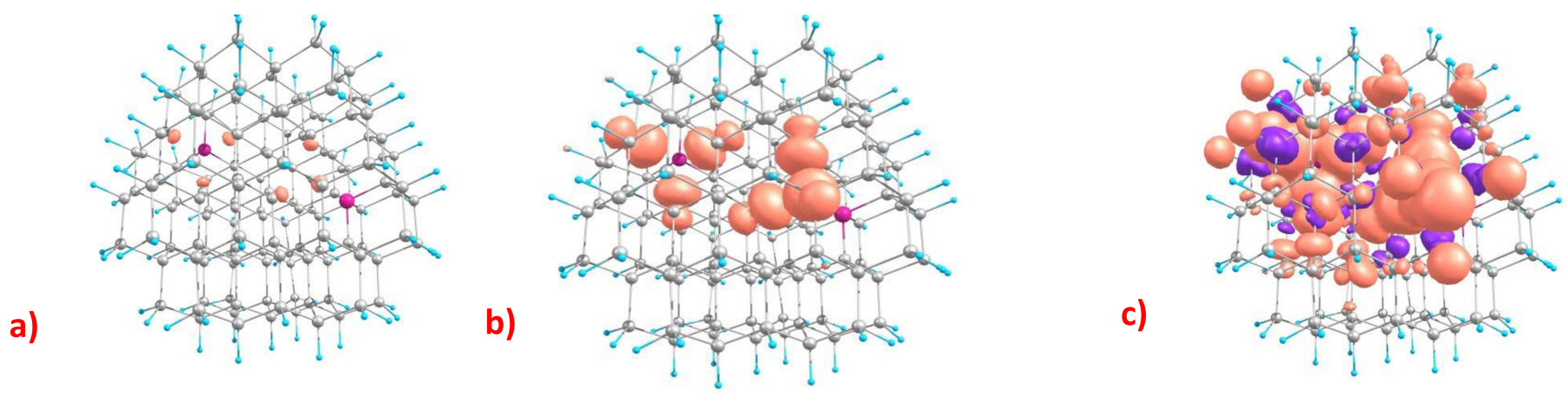
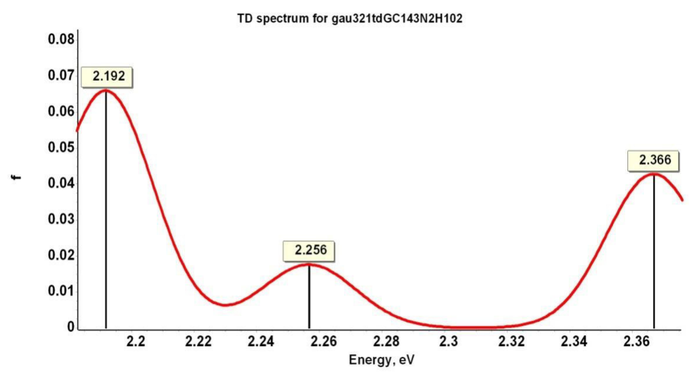
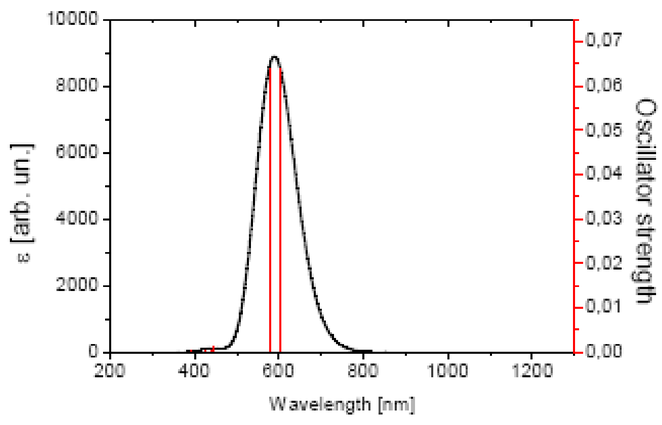


Figure 2. - Calculated isosurface of the spin density distribution of the cluster C_{143}[2NV]-H_{102}, corresponding to its values a) 0.1au^{-3}, b) 0.01au^{-3} and c) 0.001au^{-3}. Pink color corresponds to positive values of spin density, purple - negative.

It was found that the culated absorption spectrum for a cluster C_{143}[2NV]-H_{102} containing two NV centers, which was calculated demonstrates the presence of three characteristic transitions with energies in the region 2.1, 2.2 and 2.3 eV (Fig.3) , having the forces of oscillators 0.066, 0.0176 and 0.0428, respectively. The absorption spectrum for a cluster C145[NV]-H102 containing one NV center has only one maximum corresponding to 2.068 eV (Fig.4) . The resulting spectrum can be interpreted as the spectrum of a two-particle system consisting of two two-level NV centers interacting with each other and having different frequencies of optical transitions 3A → 3E due to their different position in the cluster and different orientation. Such a system is already described by four energy levels. However, unlike the case of identical and non-interacting two-level quantum systems, in this case the intermediate superpositional states A and S of the four-level system will have different energies



(Fig.3)



(Fig.4)

Basic results: Our DFT simulation confirms the possibility of developing quantum antenna consisting of two two-level emitters which are two NV centers in diamond