

# QUANTUM-CHEMICAL CALCULATIONS OF NOVEL PHOSPHORESCENT IRIIDIUM COMPLEXES

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Organic light-emitting diodes (OLEDs) are one of the most promising light sources for modern electronic devices and lighting systems [1]. OLEDs are usually made of several layers of different organic materials, each layer having a specific function. Molecular complexes containing heavy metal atom, such as Pt or Ir, are regarded as one of the most promising materials for light-emitting layer of OLEDs because of their high efficiency and wavelength tunability [2,3]. Theoretical and experimental research of such compounds is therefore crucial in development of OLED devices.

Here we present results based on quantum-chemical calculations of structural and electronic properties of bis(2-phenylbenzothiazole)-iridium-acetylacetonate ((pbt)<sub>2</sub>Ir(acac)) molecular complex and its derivatives. Atomic structure and excitation properties of the compounds are determined using several computational models; the results are compared between the models and to the experimental data. The calculations were performed using Gaussian03 and GAMESS-US programs.

The molecular structure of separate pbt ligand and the entire complex was optimized using density functional theory (DFT) with B3LYP functional and 6-31G(d) basis set. Absorption spectra of the resulting compounds were obtained using time dependent DFT (TD-DFT) method. Influence of the polarization continuum solvent model (PCM) to the calculation results was also tested; solvent effects were found to have an impact on excitation properties, while changes in molecule geometry were insignificant for these complexes. The optimized geometries were in good correspondence to the results of similar theoretical studies [4], and moreover, the absorption spectra agreed fairly well with the experimental data.

In addition to the ground-state structure (S<sub>0</sub>) geometry optimization of the lowest triplet state (T<sub>1</sub>), as well as calculations of the excited triplet states at both geometries were performed. The results indicate that the lowest-lying singlet and triplet states are both strongly linked to HOMO-LUMO electron transition between iridium atom and one of the two pbt ligands. Because of the identical ligands, the excitation spectrum in the S<sub>0</sub> geometry exhibits pairs of states with very similar differences in the excitation energies; however, this symmetry is broken in the T<sub>1</sub> geometry (Fig. 1).

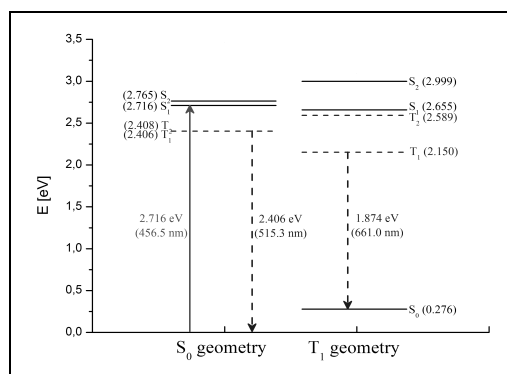


Figure 1. Lowest excited states of (pbt)<sub>2</sub>Ir(acac) complex in the S<sub>0</sub> and T<sub>1</sub> geometries

Geometry optimization and excited states calculations of the extended ligand with additional carbazole groups and the resulting Ir complex were performed using the DFT method. It is shown that wavefunction character in the lowest excited states is not affected by addition of the charge transporting groups.

Finally, the interaction between singlet and triplet energy states was explored. The interaction strength was evaluated from the splitting of energy states caused by spin-orbit coupling. Additional concern for the accuracy of computational results is determined by relatively heavy sulfur atom present in the pbt ligand. To assess the possible effect of the S atom, additional optimization was performed using model core potential approach with triple-zeta valence basis set (MCP-TZP, as implemented in GAMESS-US). Resulting geometries and absorption spectra were compared to the experiment results.

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