Ambipolar concave PAHs with manifold, precisely arranged, nitrogen dopants. Unprecedented class of efficient OLED emitters.

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Motivation: The use of structurally modified polycyclic aromatic hydrocarbons (PAHs) with a non-planar bowl structure, containing heteroatoms precisely embedded inside the organic scaffold, allows for strict control of their physicochemical properties. The most frequently presented, containing one nitrogen atom in their structure, are electron-rich p-type semiconductors. However, their structure does not allow for further synthetic fine-tuning of optical properties, which is necessary for potential application as new third generation OLED emitters. In the last two years, a new concept of multi-resonance OLED emitters based on planar organic structures comprising symmetrically arranged nitrogen atoms in 1,3/1,4 positions has been introduced. While they are good blue light emitters, due to the low availability of potential building blocks and the need to fill the periphery of molecules with units preventing their aggregation, it is not possible to introduce units reducing the HOMO-LUMO energy gap responsible for the color of emissions. Introducing the possibility of multiresonance response to non-planar bowl-type skeletons will allow the maximum use of the advantages of both planarity disturbance (TADF, increased solubility) and multiresonance (increased emission efficiency).

The aim of research: In this project, carried out by the **first Polish organic optoelectronics consortium**, we will aim at computer-aided synthesis of new, highly symmetrical dyes based on (i) 1,4/ 1,3/1,2- dinitrogen containing bowl-shaped molecules bearing fully conjugated (ii) electron-deficient fragments in their structure. The implementation of such a topology (i) is envisaged to eliminate overlapping of the HOMO-LUMO orbitals, which is crucial to achieving the efficient TADF emission in OLED devices. Moreover, the proposed structurization would allow to precisely adjust the electronic character, not only of the fully conjugated electron-deficient units (a), but would, for the first time, open a way of introducing additional electron-rich groups (b), so that using both (a / b) **would enable not only to freely tune the emission range, but to simultaneously improve all optoelectronic properties at once**, i.e. fluorescence quantum efficiency (PLQY) and external emission quantum efficiency (EQE), not just a single as has been the case so far. The project carried out by our consortium will follow a stepwise approach between different sections, including computer-aided molecular design to allow for the initial optimization of the geometry and electronic properties of the proposed dyes (head: Prof. A. Kubas). Parallel to the theoretical research, we will start synthetic work, which is the core of this project, including the preparation of compounds: with different degrees of bowl size, location of resonant nitrogen atoms, regioisomeric alignment of electron-deficient substituents, and arrangement of electron-rich units (head: Dr. M. Lindner). Subsequently, we will perform photophysical characterization of the obtained target dyes in solution and solid state, which will be next utilized as emission layers in MR TADF OLED devices (head: Prof. P. Data).

Expected impact of the research project: Our strategy of fine-tuning electronic structure of bowl-shaped organic architectures is appealing from viewpoint of both fundamental and even applied research. This is because structurally aesthetic molecules dyes with tailored resonant nitrogen atoms and properly implemented electron-deficient subunits can be offered as a new highly efficient OLEDs and shall thus cause sudden breakthrough in a new domain of nitrogen-based multiresonant emitters.

Proposed research also covers the aspect of **sustainable management and saving of the energy** by using alternative sources to commercially available materials. This is relevant to the lighting and displaying technologies in which, expensive and environmentally unfriendly, noble metal complexes are being used. Future replacement by the organic emitters, that are easier to be utilized and thus less hazardous, is supposed to meet the prerequisites of the "**European GreenDeal** " which was released to the public by the European Commission which is one of the most important program realized by European commonwealth. Our approach is also in a line with the importance of a better energy management which was rose during recent **World Economic Forum in Davos** (https://www.weforum.org/agenda/2022/01/iea-energy-efficiency-worlds-first-fuel-net-zero)