

行政院國家科學委員會專題研究計畫 期中進度報告

當前一些重要化學反應之快速光譜，光譜動力學探討(2/3) 期中進度報告(精簡版)

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九十五年度[當前一些重要化學反應之快速光譜, 光譜動力學探討] 成果報告以及經費變更

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一. 計劃成果報告

立基在光物理以及分子結構的相關性原理上, 我們最近已經由設計與合成, 在一些嶄新的電子及質子轉移化合物上大放異彩。首先, 我們根據 HOMO-LUMO 電子轉移的基礎架構, 設計出了全世界第一個類螢光蛋白官能基的發光團。不同於光蛋白發色團需要 3 度空間(3-dimentional), 鋼硬構體 (rigidity)以及水分子及胺基酸的正確排列才能進行電子轉移激發質子轉移, 此 ortho-異構物藉由本身的七元環內氫鍵即能進行電子轉移感應質子轉移的反應。我們進而用 fluorescence up-conversion 以及 transient absorption 加上 deuterium isotope effect, 結論出激發態質子轉移是沒有能障限制, 甚至是一個 coherent motion. 目前的進度是藉由各種化學修飾可以微調波長下, 其一系列衍生物的基礎及應用研究將是無遠弗屆。另外也是藉由化學設計及合成原理, 我們已經發展出一系列的電子/質子轉移偶合新化合物, 且致力於其基礎反應動力學以及應用上的研究。在基礎的研究上我們已經經由強耦合以及若耦合的理論基礎來闡釋各種不同的實驗結果, 並試圖讓我們的結果能以一統化的理論來歸納。這部份的成果也相當的傑出。日前已經獲 Acc. Chem. Res.的邀稿在寫一個相關的 review 論文。在應用方面我們也藉由電子/質子轉移的概念成功的在雷射染料, 有機發光元件以及太陽能電池的修飾上有相當傑出的成果。我們對於耦合機制瓶頸克服之道已設計出一系列新的化合物, 其發展非常的順遂, 但這些系統目前尚未達到最適切 (optimized)的境界。我們的下一個目標是設計一個全新的系統, 使其在第一激發態沒有任何激發態電子轉移的發生, 而是在質子轉移後伴隨電子轉移的反應。目前的進展相當順利, 應在半年之內有所突破。據此, 我們深具信心可以進一步、更透徹的來了解激發態電子及質子轉移的機制。子計劃裏我們提出並在這一年已經以合成方式設計出不同架

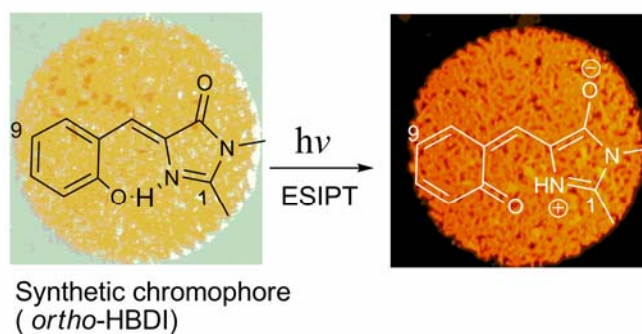
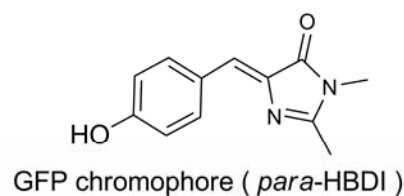
橋所連結的雙色團、甚至多重色團來研究激發態電子轉移的有效機制。我們的遠程目標: 雙色團架構配以不同長度, 構型的架橋, 甚至利用氫鍵的多樣化(flexibility and versatility)來研究電子轉移的適合參數, 目前正在進行中。我們希冀能夠設計多重色團, 經由不同架橋的組合達到設計分子導線甚至光致電元件的最佳組合。我們也將在新的實驗中以飛秒雷射結合螢光合頻以及瞬態吸收光譜技術來做緩解動力學, 再用 IR 偵測系統來探測其所伴隨的結構變化。我們在這方面相關系統的設置已頗具規模, 未來相信可以接受整合計畫相關的更高困難實驗挑戰。

受限於 space, 我們在這只列三個成果實例。如果有興趣了解更多的 details 請參閱我們所列的 references 中所打星號部份。

三. 執行進度

A. ortho-GFP Synthetic Chromophore; Excited State Intramolecular Proton Transfer via a Seven-membered Ring Hydrogen Bonding System

In this study, a structural isomer of the core emission chromophore 4-(4-hydroxybenzylidene)-1,2-dimethyl-1H-imidazol-5(4H)-one (para-HBDI) in green fluorescence protein (GFP), namely ortho-HBDI, has been synthesized. ortho-HBDI possesses a seven-membered ring hydrogen bond, from which the excited-state intramolecular proton transfer (ESIPT) takes place, resulting in a remarkable proton transfer tautomer emission of ~ 605 nm in organic

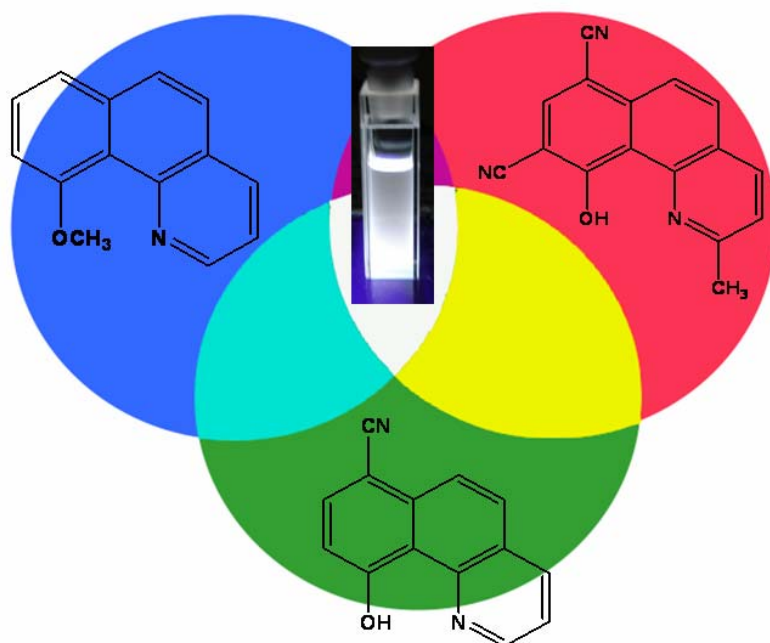
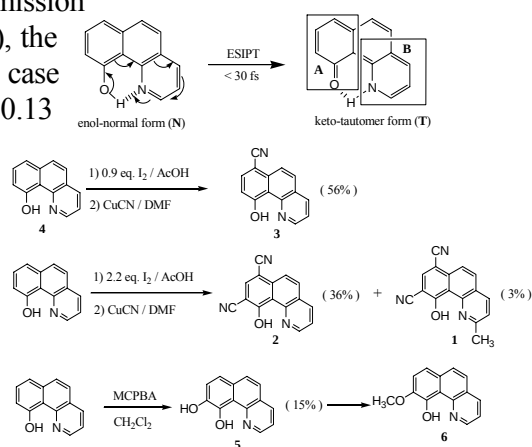


solvents. Both deuterium isotope experiments and theoretical approaches point to an essentially barrierless potential energy surface along the ESIPT reaction. ESIPT also takes place in the solid film of ortho-HBDI, resulting in a ~ 595 nm tautomer emission with a quantum yield as high as 0.4. In sharp contrast, unless in the presence of GFP under a hydrogen-bond network, the GFP free para-HBDI gives nearly no emission in both solution and solid. While the bioactivity of ortho-HBDI is pending further

exploration, its future chemical derivation is versatile. In an ongoing project, via the functionalization at C(1) and C(9) positions, the proton-transfer emission can be tuned to a broad range from green to deep red with high emission efficiency in solid film, generating a new series of isomers of para-HBDI with remarkable excited state intramolecular proton transfer properties.

B. Extensive spectral tuning of the proton transfer emission from 550 to 675 nm via a rational derivatization of 10-hydroxybenzo[h]-quinoline.

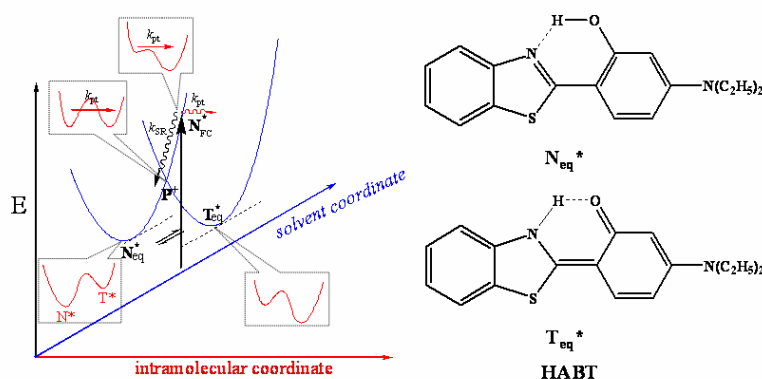
In this study, on the basis of a ESIPT system HBQ, we have systematically synthesized a new series of derivatives, such that the proton-transfer emission can be extensively tuned from 550 nm (**1**) to ~680 nm (**6**), the emission yield of which obeys the energy gap law. For the case of **1-3**, the emission quantum yield was measured to be > 0.13 in ethyl acetate, and ASE was readily observed, generating a new family of proton transfer fluorescence dyes. Future applications of this series of ESIPT molecules can be greatly expanded. For example, via mixing e.g. **1** ($\lambda_{\text{em}} \sim 550$ nm), **3** ($\lambda_{\text{em}} \sim 605$ nm) and the methoxylated derivative of **4**, in which ESIPT is prohibited (emission $\lambda_{\text{max}} \sim 420$ nm, $\Phi \sim 0.3$ in ethyl acetate),¹⁴ a qualitative white light generation can be achieved with a regular UV lamp (366 nm).



Via mixing compounds **1**, **3** and **4a**, a qualitative white light generation can be achieved in ethyl acetate with a regular UV lamp (366 nm)

C. Spectroscopy and Femtosecond Dynamics on 2-(2'-Hydroxy-4'-diethylaminophenyl)benzothiazole; The Role of Solvent Polarity in the Excited-State Proton Transfer Reaction

Detailed insights into the excited state enol (N^*)-keto(T^*) intramolecular proton transfer (ESIPT) reaction in 2-(2'-hydroxy-4'-diethylaminophenyl)benzothiazole (**HABT**) have been investigated via steady state and femtosecond fluorescence up-conversion approaches. In cyclohexane, in contrast to the ultrafast rate of ESIPT for the parent 2-(2'-hydroxyphenyl) benzothiazole ($>2.9 \pm 0.3 \times 10^{13} \text{ s}^{-1}$), **HABT** undergoes a relatively slow rate ($\sim 5.4 \pm 0.5 \times 10^{11} \text{ s}^{-1}$) of ESIPT. In polar, aprotic solvents competitive rate of proton transfer and rate of solvent relaxation were resolved in the early dynamics. After reaching the solvation equilibrium in the normal excited state (N_{eq}^*), ESIPT takes place with an appreciable barrier. The results also show $N_{eq}^*(\text{enol}) \leftrightarrow T_{eq}^*(\text{keto})$ equilibrium, which shifts toward N_{eq}^* as the solvent polarity increases. Temperature dependent relaxation dynamics further resolved a solvent induced barrier of 2.12 kcal/mol for the forward reaction in CH_2Cl_2 . The observed spectroscopy and dynamics are rationalized by a significant difference in dipole moment between N_{eq}^* and T_{eq}^* , while the dipolar vector for the enol form in the ground state (N) is in between that of N_{eq}^* and T_{eq}^* . Upon $N \rightarrow N^*$ Franck Condon excitation, ESIPT is energetically favorable, and its rate is competitive with the solvation relaxation process. Upon reaching equilibrium configurations N_{eq}^* and T_{eq}^* , forward and/or backward ESIPT takes place with an appreciable solvent polarity induced barrier due to differences in polarization equilibrium between N_{eq}^* and T_{eq}^* .



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- 149 “Design and Synthesis of Ir(III)-Azacrown Complex: Application as a Highly Sensitive Metal Cation Phosphorescence Sensor Mei-Lin Ho,^a Fu-Ming Hwang,^b Pei-Nung Chen,^b Ya-Hui Hu,^a Yi-Ming Cheng,^a Kung-Shih Chen,^a Gene-Hsiang Lee,^a Yun Chi,^{b,*} and Pi-Tai Chou^{a,*}“**Org. Biomol. Chem. 2006, 4, 98. (2.6)**

這次芝加哥第 233 屆 ACS 會議，我和我的博士班學生何美霖以及博士後鄭宜明收獲相當大。我們主要的 focus 是在最新、最進步、最有前途的幾個研究領域，分為技術層面以及研究主題兩方面。當然主要的訴求是和物化、材料方面和我們實驗室研究相關的題材。其中在技術層次方面最讓我們驚異的是 2D-IR 的發展。2D-IR 在時間解析方面比之於 2D-NMR 或其他的 structural resolved spectroscopy 都來的短，且在官能基的資訊方面有相當大的優越程度。目前大概發展了 6~7 年，都屬於國際頂尖物化實驗室在發展。目前已跨小分子而至生化分子發展，未來應用可能無遠弗屆，但這方面所涉及的原理及技術相當複雜，需要跨領域的合作，加上目前商業上仍沒有 turn-key 系統，在國內發展的話可能還需要一些時間，也只怕建好後後續沒有資質好的學生能夠維持。

在研究主題方面仍是以能源相關研究最熱門，尤其是太陽能電池相關領域演講場場爆滿。這方面相信也是國內最應發展的方向之一。我們也給了兩個不同主題的 poster section 以及和相關領域的研究者交流。綜合論之，這是一趟收獲豐碩之旅，無愧於國科會輔助。