

# Hydrogen activation by frustrated Lewis pairs: Comparison of reactivity models

Imre Pápai, Tibor András Rokob, Imre Bakó, András Stirling, Andrea Hamza

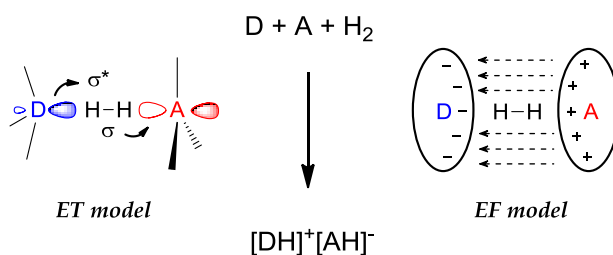
Laboratory of Theoretical Chemistry, Institute of Organic Chemistry,  
Research Centre for Natural Sciences, Hungarian Academy of Sciences, Hungary

H-1025 Budapest, Pusztaszeri út 59-67, E-mail: papai.imre@ttk.mta.hu

Lewis acid–base pairs with bulky substituents, the so-called frustrated Lewis pairs (FLPs), have recently shown to be reactive species towards small molecules. For instance, they can easily split dihydrogen into a proton/hydride pair and act as transition-metal-free catalysts for the hydrogenation of unsaturated compounds.

Very limited experimental information is available regarding the mechanism of these reactions; however, computational studies provided valuable mechanistic insight [1]. From these investigations, two alternative reactivity models have emerged to interpret the facile FLP-mediated heterolytic cleavage of H<sub>2</sub>. Both models assume that the reaction takes place via reactive intermediates involving properly oriented Lewis donor (D) and acceptor (A) partners, but they differ conceptually in defining the basic interactions in the activation process. In the electron transfer (ET) model, the cleavage of H<sub>2</sub> is related to synergistic electron donations incorporating both active centers of the FLP and the bridging hydrogen, whereas the electric field (EF) model suggests that the heterolytic bond cleavage occurs as a result of electrostatic polarization by the strong field being present between the donor and acceptor partners (see scheme below).

In this contribution, the two models will be paralleled in terms of their abilities to account for the computed reaction characteristics as well as for the observed reactivities [2].



[1] Rokob, T. A., Pápai, I. *Top. Curr. Chem.*, 332:157-212, 2013.

[2] Rokob, T. A., Bakó, I., Stirling, A., Hamza, A., Pápai, I. *J. Am. Chem. Soc.*, 135:4425-4437, 2013.