Organic "diradicals" are important for the synthetic chemist who wants to exploit them as precursors of target molecules, for the mechanistic chemist who seeks to unravel reaction pathways, for the quantitative theoretician who is anxious to test different computational schemes on such molecules because of the formalistic intricacies involved, and for the qualitative theoretician who seeks to understand how these species are bound. Specialists of the latter two types most often adhere to MO theory and they discuss the electronic properties of "diradicals" in the following way: They depart from Hückel MO theory and point out why neglect of interelectronic repulsion renders it inapplicable to problems involving "diradicals". Then, the discussion shifts to the SCF-MO level and various formal drawbacks and resulting pitfalls are recognized. Finally, one is forced to examine the problem at the SCF-MO-CI level, something which guarantees that the potential audience of the paper is exponentially reduced and that the ensuing discussion is rendered cumbersome and lengthy. In a recent work,¹ we advanced the argument that qualitative Valence Bond theory has the formal correctness and conceptual clarity which can allow one to dispense with problems which are hard to deal with within the MO theoretical framework in the space of a paragraph or two. In particular, in treating homonuclear systems involving relatively weak π bonds, one can use the Approximate Heitler-London (AHL) theory outlined in the original monograph. Furthermore, if the problem does not involve parity distinctions, AHL theory can be substituted by the nearly half-century old Diatomics in Molecules (DIM) approach introduced by Eyring.²-¹² According to this brand of theory, the relative stability of isomers is mainly due to overlap bonding or overlap antibonding and the DIM eigenstates are excellent approximations of the corresponding SCF-MO-CI eigenstates, under the conditions stated above. Noting that cyclobutadiene(CB) and trimethylenemethane(TMM) have attracted a large amount
of attention in the last ten years and desiring to show exactly how trivial most of the associated problems are when looked from the standpoint of qualitative VB theory (in this case DIM theory), we have calculated the ground state energies of these controversial organic diradicals by using procedures known since the 1930's. The results are given in Table 1 and they constitute direct answers to many often-discussed problems. Since the coulomb integral Q is approximately invariant, relative energies depend on the number and signs of the exchange terms, T, each of which describes a two-electron bond (positive sign preceding T) or a two-electron antibond (negative sign preceding T). The following conclusions are the same as those arrived at by ab initio SCF-MO-CI computations after considerable controversy generated by less than adequate monodeterminantal SCF-MO calculations:

a. Singlet CB is more stable than triplet CB.

b. Triplet TMM is more stable than singlet TMM.

c. Singlet TMM is less stable than allyl plus pi so it must have a perpendicular conformation.

d. The electronic structures of the various species extracted from laborious SCF-MO-CI computations are essentially those shown in Table 1.

If we replace DIM by AHL theory so that the effect of parity on stereoselection can be "seen", one obtains the Woodward-Hoffmann predictions that Möbius CB will have lower energy than Hückel CB and, analogously, Möbius TMM will be more stable than Hückel TMM. That is to say, TMM is predicted to close thermally to methylene cyclopropane by conrotation.

Finally, I have added to the Table the predictions of DIM theory for the degenerate isomerization of 3 H₂ via an H₆ hexagon, a model of an "allowed" reaction. We must conclude that barriers exist in "allowed" reactions because of interfragmental overlap repulsion and that concepts like N-electron-N-orbital-N-center homoconjugation are invalid. It is then clear that many of the current