Reformulating Time-Dependent Density Functional Theory (TDDFT) with Non-orthogonal Localized Molecular Orbitals (supplementary materials)

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I. THE "ON THE FLY" RE-LOCALIZATION

The re-localization operation is another important point in this work, making NOLMOs sparse in the whole of propagation process. The sparsity of NOLMOs's distribution of molecule $C_{60}H_{122}$ in initial time was shown in Fig.1, which was very sparse in AO representation. However, it would spread along time propagation as shown in Fig.2, increasing scaling of computations. However, with our "on the fly" re-localization technique, the new sparse NOLMOs' distribution could be also re-generated, as shown in Fig.3. This characteristic also exists in conjugated systems.

Compared with NOLMO, the CMO is very delocalized, as shown in Fig.4.

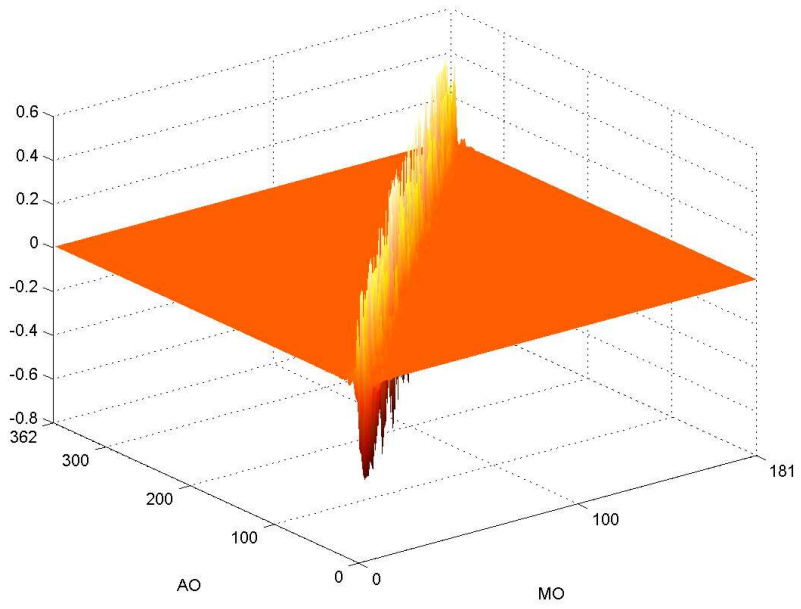

Figure 1: The real part of NOLMOs in the initial step.

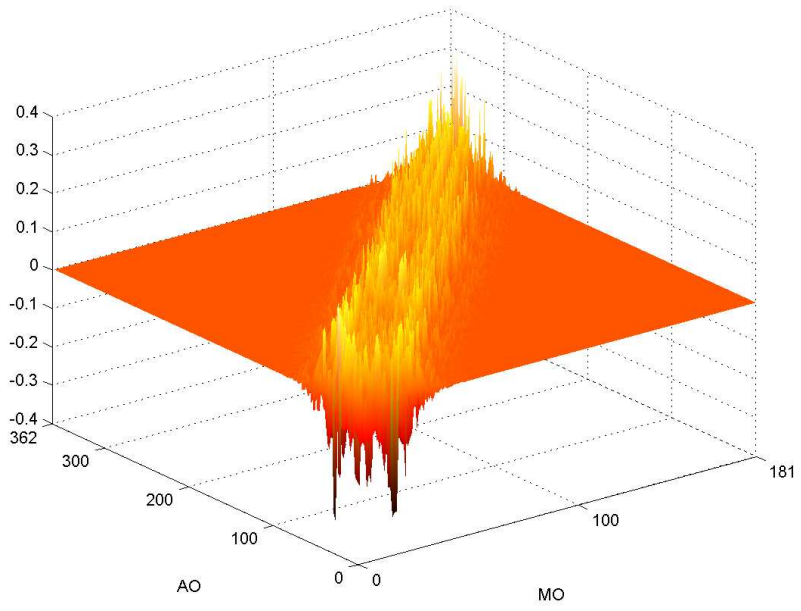

Figure 2: The sparsity of NOLMOs' distribution after 50 au propagation.

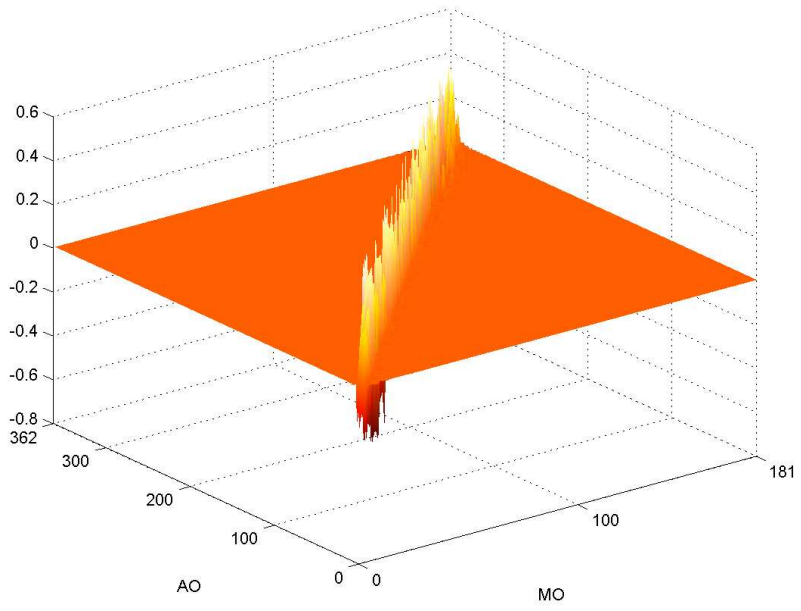

Figure 3: The sparsity of NOLMOs' distribution after a re-localization operation at 50 au.

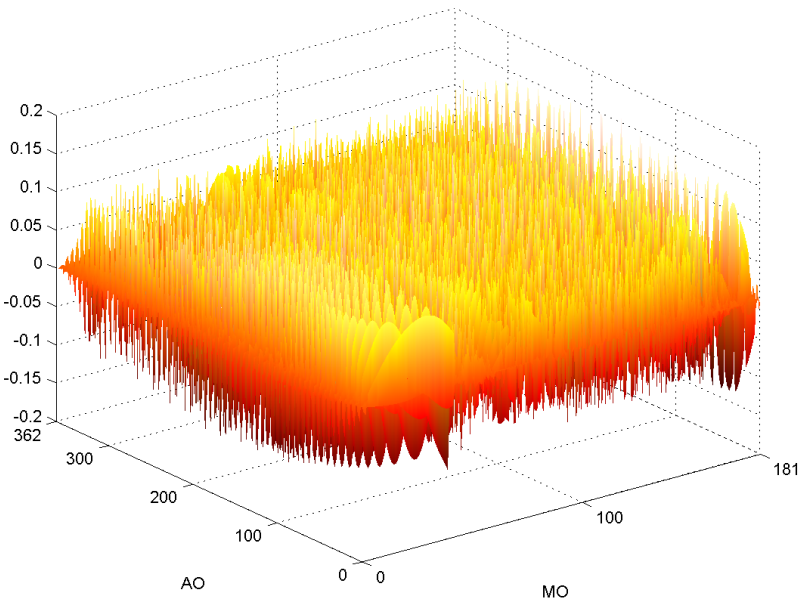

Figure 4: The distribution of CMOs in AO space.