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Generalized Elongation 法の開発と応用

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The elongation method [1-4] has been developed since 1990s in our laboratory for efficiently calculating electronic structures of polymers. The original idea comes from experimental polymerization/copolymerization procedures. Contrast to other approximation methods for large systems, the elongation is fully variational. It is confirmed that the total energies obtained by this method are in excellent agreement to those by the conventional method. The error of the elongation method is within 10^{-9} a.u./atom [5-7]. The elongation method has been applied to various periodic and/or aperiodic quasi-one-dimensional systems [8-10] for the electric and optical properties.

However, the original implementation of the elongation method was restricted only to quasi-one-dimensional systems. It has been questioned whether it is applicable for three-dimensional systems. In this presentation, a generalized elongation method will be presented for any dimensional systems. Fig. 1 is a schematic show of the generation elongation (G-ELG) method. The blue circles are frozen units while the red circles are active units. As system is elongated, the active region (within the pink circle) can be approaching to the previously frozen units, leads to strong interaction between them. The G-ELG method recognizes this interaction and first re-activates those frozen units. After the elongation SCF converged, the CMOs will be re-localized so that the next elongation step will be continued without increasing the size of the interactive region. By repeating this procedure, any random with any-dimensional system can be generated by the G-ELG method.

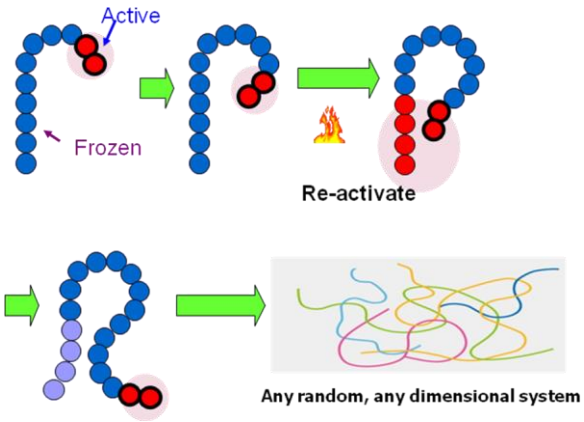

Figure 1. Schematic show of the generalized elongation method, the pink region is the interactive region.

Fig.2 is the 3-methyl-4-nitropyridine-1-oxyde (POM) molecular crystal structure. Left panel is the unit cell and the right panel shows how the routine of the elongation method to form these eight units cluster. The G-ELG method results, as shown in Table I, demonstrate that the high accuracy can be achieved if some frozen units are included in

the active region. It shows that the G-ELG method is applicable to three-dimensional systems.

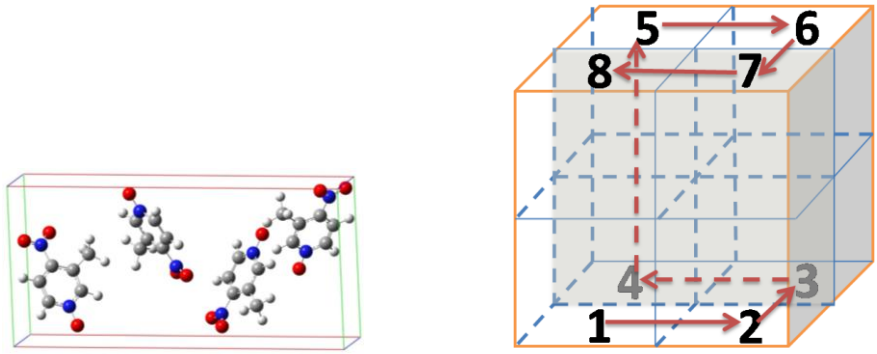

Figure 2. The 3-methyl-4-nitropyridine-1-oxyde (POM) molecular crystal. Left panel is the unit cell and the right panel shows how the routine of the elongation method to for these eight units cluster.

TABLE I. Energies between the conventional and elongation calculations for POM molecular crystal. All the energies are given in a.u. and the number of activated units means the originally frozen units are activated in the current elongation step.

N	CNV	Old-ELG	△E	G-ELG	△E	#of activated
3	-6679.879802	-6679.879802	-1.99179E-10	-6679.879802	1.99179E-10	0
4	-8906.518042	-8906.479395	0.038647476	-8906.518042	-1.00044E-10	1
5	-11133.13369	-11133.04172	0.091965002	-11133.13369	2.66200E-06	1
6	-13359.74954	-13359.53218	0.217361617	-13359.74954	2.54960E-06	2
7	-15586.38538	-15586.06323	0.322143759	-15586.38537	1.06245E-05	2
8	-17813.02496	-17812.50852	0.516442567	-17813.02496	-2.00089E-10	5

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