Abstract Submitted
for the MAR10 Meeting of
The American Physical Society

Periodic image errors in the electronic hyperpolarizabilities Y. TAKIMOTO, ISSP, the University of Tokyo, M. OTANI, AIST Tsukuba, O. SUGINO, ISSP, the University of Tokyo — Second-order nonlinear optical properties of large organic nonlinear optical (NLO) molecules have attracted increasing attention, and static and dynamical hyperpolarizabilities are now important target of the ab-initio theoretical studies. In the DFT and TDDFT, the Poisson equation is commonly solved with the periodic boundary condition (PBC) to reduce the computational cost, but it produces serious the periodic image error due to large dipole moment of the NLO molecules. Here, we show that a Green's function technique, i.e., effective screening medium (ESM) method of Otani and Sugino, provides an efficient way of overcoming this problem. The ESM method allows to apply an external electric field to the molecule by changing the boundary condition of the Poisson equation. Moreover, the method requires the computational cell just enough to contain the electron density, although PBC calculation requires the dipole-interaction tail to sufficiently small at the cell boundary. Other correction methods such as cut-off or multipole method also requires larger, say, doubled cell. The ESM method has a further merit of using various boundary conditions to model surrounding vacuum, metallic as well as dielectric medium. This suggests a future possibility of incorporating the solvent effects into the theoretical study. The ESM method thus would stimulate further studies of a large photonic molecule.

Yoshinari Takimoto
ISSP, the University of Tokyo

Date submitted: 20 Nov 2009 Electronic form version 1.4