Ordering of octupolar molecules for nonlinear optics applications: searching for a robust scenario

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We evaluate various approaches for effective electric field poling of interacting octupolar molecules[1], depicted in Fig. 1ab. A two-dimensional lattice model, that includes important features of poling, is studied using the complementary approaches of analytical methods in statistical mechanics and Monte Carlo simulations. Quantum chemistry is used to check some parameters and assumptions of the model. We also compare the experimentally accessible nonlinear-optical properties resulting from each scenario.

The inhomogeneous poling field is imparted by a system of point charges of adequate symmetry, see Fig. 1c. The possibility of practical construction of the octupoling cell is discussed by taking into account electric breakdown, mechanical strength, heat dissipation etc. Because a very weak degree of acentric order is sufficient for nonlinear detection, and very strong electric fields are applicable without electric breakdown in certain polymeric systems, the revised poling criteria are improved by four orders of magnitude w.r.t. the earlier paper[2], leading to required temperature of a few Kelvins.

We discuss another scenario: dipoling of octupolar molecules bearing small dipole. We strictly develop the possibility of obtaining octupolar order by strong homogeneous field poling at Nitrogen temperature, although the octupolar order was previously predicted to vanish in this case[3] using high temperature approximation.

Finally, the electrostatic intermolecular interactions are considered, see Fig. 2, in hope for a decisive increase of required temperature. A lattice system of 1000 interacting octupoles that can take any 3D orientation is considered. It is shown to stay in a plane and perfectly ordered up to 80 K. Moreover, if the molecules are forced to stay in a plane (for example on a substrate) the effective temperature can approach room temperatures.

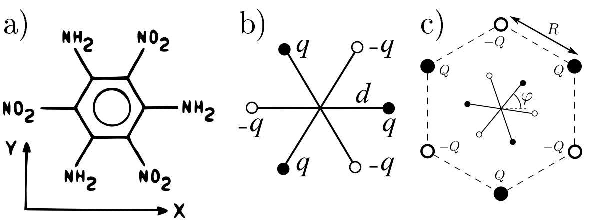

Figure 1: (a) The prototype of a small octupolar molecule. (b) The model six-arm octupole. (c) The model octupole in a poling cell of size $R \approx 60$ nm consisting of point charges $\pm Q$.

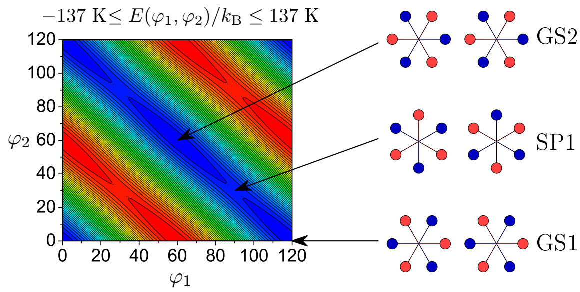

Figure 2: Potential energy surface $E(\varphi_1, \varphi_2)$ for a pair of electrostatically interacting octupoles at fixed distance $l = 5d$ between their centers.

[1] J. Zyss, Nonl. Opt. Quant. Opt. **43**, 97 (2012).
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[3] S. Brasselet, J. Zyss, J. Opt. Soc. Am. B **15**, 257 (1998).