

Effects of PEG addition on particle-particle interactions in crystal growth of DNA functionalized nanoparticles

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Due to the programmability of DNA base sequences, DNA-functionalized nanoparticles (DNA-NPs) can be utilized as to self-assemble into various types of 3D nanoparticle superlattices [1]. However, it is not fully understood how their behavior in the solution, such as particle-particle interactions, affect crystal nucleation and growth, and consequently it is difficult to produce large high-quality single crystals. Here, we report on the new effect of PEG addition on crystal growth of DNA-NPs and show that the crystal size can be increased by the excluded volume effect of PEG [2] (Fig. 1).

Au nanoparticles were, respectively functionalized with a pair of complementary sequenced thiolated-DNA strands. We heated solution up to 65°C and then slowly cooled back to 25°C. The crystal structures of DNA-functionalized gold nanoparticles (DNA-AuNPs) superlattices were analyzed by small angle X-ray scattering (SAXS). The crystal shapes were observed by scanning electron microscopy (SEM).

DNA-AuNP crystals grew larger when PEG with molecular weights of 1000-1500 were added to the crystallization solutions than when no PEG was added (Fig.2). In solvents containing high concentrations of PEG, DNA-AuNPs designed to be bcc structure tended to crystallize into a stable fcc structure. (Fig.3). The change in crystal structure is thought to be due to inter-particle forces [3].

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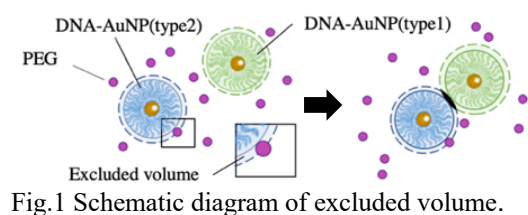


Fig.1 Schematic diagram of excluded volume.

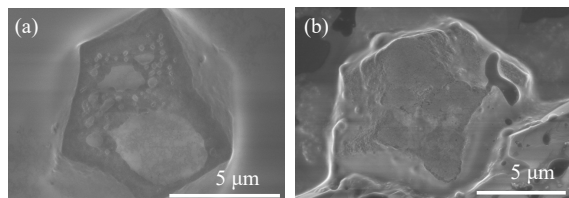


Fig.2 SEM images of DNA-AuNP crystals.
(a)PEG-free solution. (b)PEG1500 2wt.% solution.

References

- [1] Macfarlane RJ et al. *Science* 2011;334 (6053):204-208.
- [2] Shin J et al. *The Journal of Physical Chemistry B* 2012;116 (45):13396-13402.
- [3] Song M et al. *Langmuir* 2018; 34 (3):991–998.

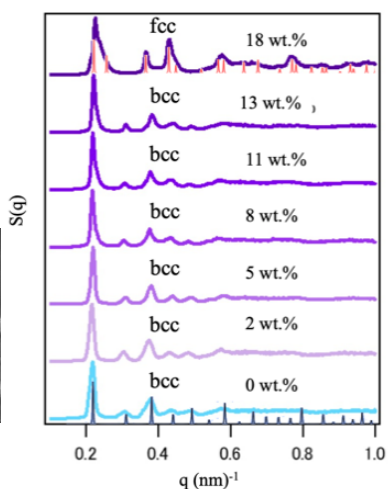


Fig.3: SAXS profiles of DNA-AuNP crystals (0.5 M NaCl, PEG1000) $q=(4\pi/\lambda) \sin\theta$ (scattering angle, 2θ). The structure of crystal in solution containing 0~13 wt.% PEG is bcc structure. The structure of crystal in solution containing 18 wt.% PEG is fcc structure.