Supporting information

Crystallization of Gd_2O_3 nanoparticles: evolution of the microstructure *via* electron-beam manipulation

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Table S1. Statistics analyses of amorphous NPs in four areas.

	Area 1	Area 2	Area 3	Area 4
Total No. of NPs	90	73	92	72
No. of crystalline *cubic*-$NaGdF_4$	71	55	70	59
No. of amorphous NPs	19	18	22	13
Proportion of amorphous NPs (%)	0.21	0.25	0.24	0.18

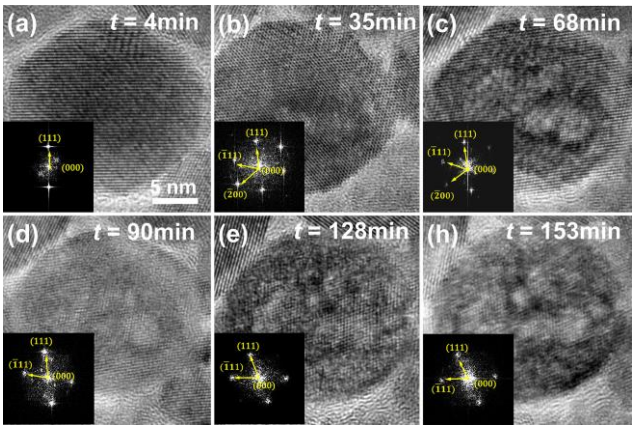

Fig. S1 Time-resolved structure evolution for **Particle 3**.

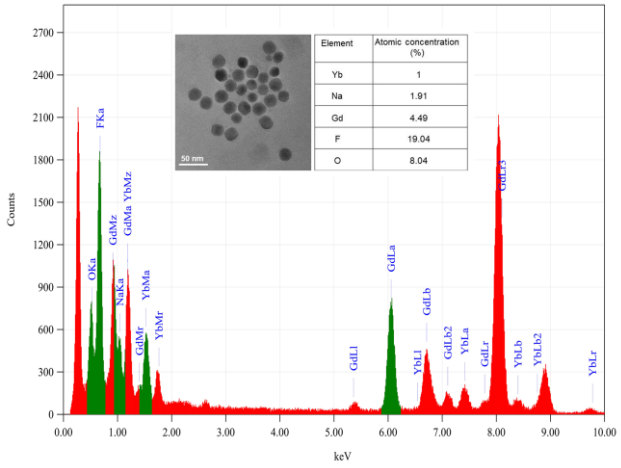

Fig. S2 EDS spectrum and analysis of as-synthesized $NaGdF_4$:Yb,Er NPs deposited on Si-based SiN_x film.

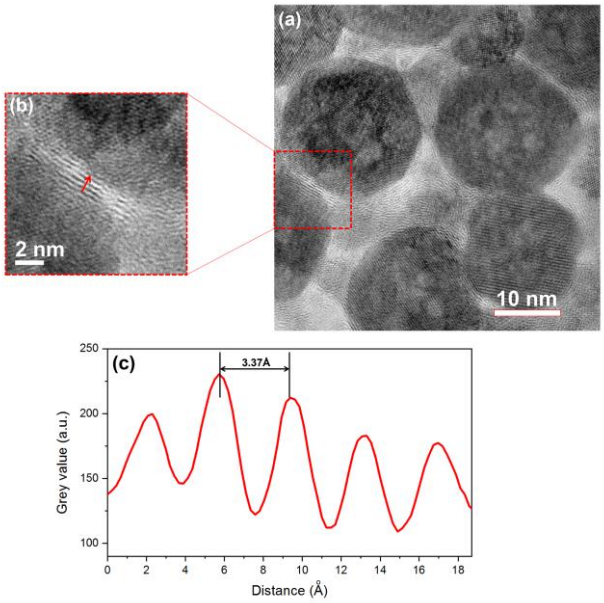

Fig. S3 (a) Formation of multilayer graphene on the surface of nanoparticles and in their surroundings after *e*-beam irradiation for 112 min. (b) The magnified TEM image of the red dashed square in (a), which shows clearly the lattice fringes of graphene with *d*-spacing of 3.37Å, corresponding to (0002) plane of graphite. (c) The profile of the line scan of the red arrow in (b), which shows the averaged *d*-spacing is 3.37 Å.

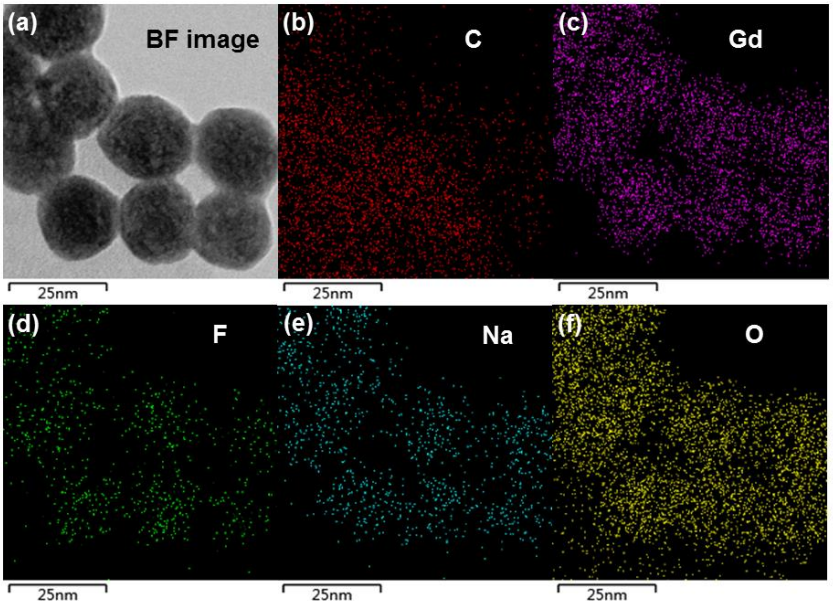

Fig. S4 STEM-EDS mapping of the NPs after 125 min irradiation with beam current of 60 pA cm^{-2}. The sample was deposited on Si-based SiN_x film to avoid the disturbance of carbon film. The mapping results exhibit the co-existence of C, Gd, Na, F and O atoms,

giving clear evidence of the formation of carbon species.