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Supporting Information for

"Synthesis-driven, structure-dependent optical behavior in phase-tunable $NaYF_4$:Yb, Er-based motifs and associated heterostructures"

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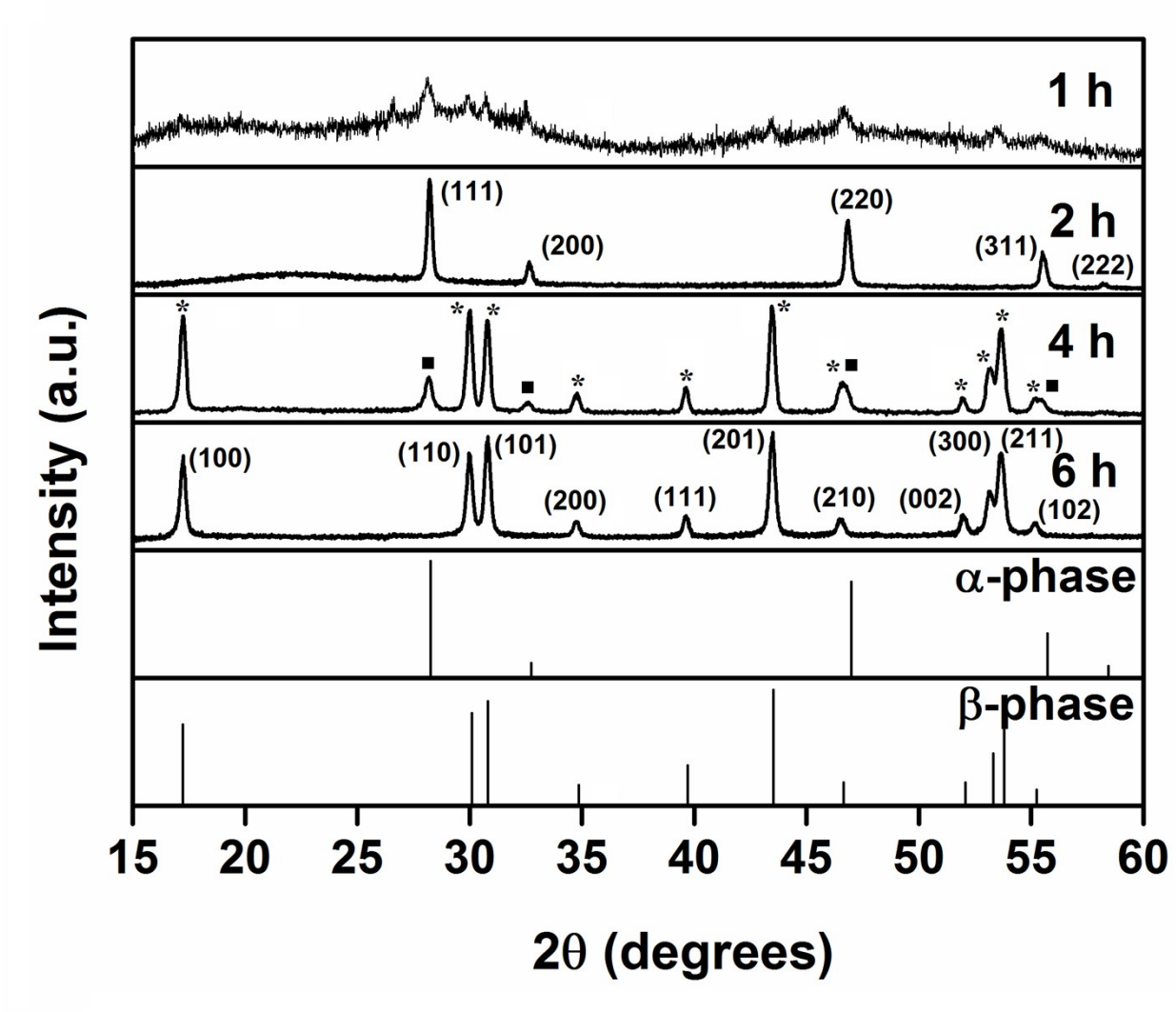

Figure S1. XRD patterns of as-prepared $NaYF_4$ samples, generated after *reaction times* of 1 h, 2 h, 4 h, and 6 h, respectively. The 'stars' designate hexagonal-related facets, while the 'squares' correlate with cubic-related facets. The reaction temperature has been fixed at 100°C, while the ammonia concentration has been set at 0.2 M, for all four of the samples processed herein.

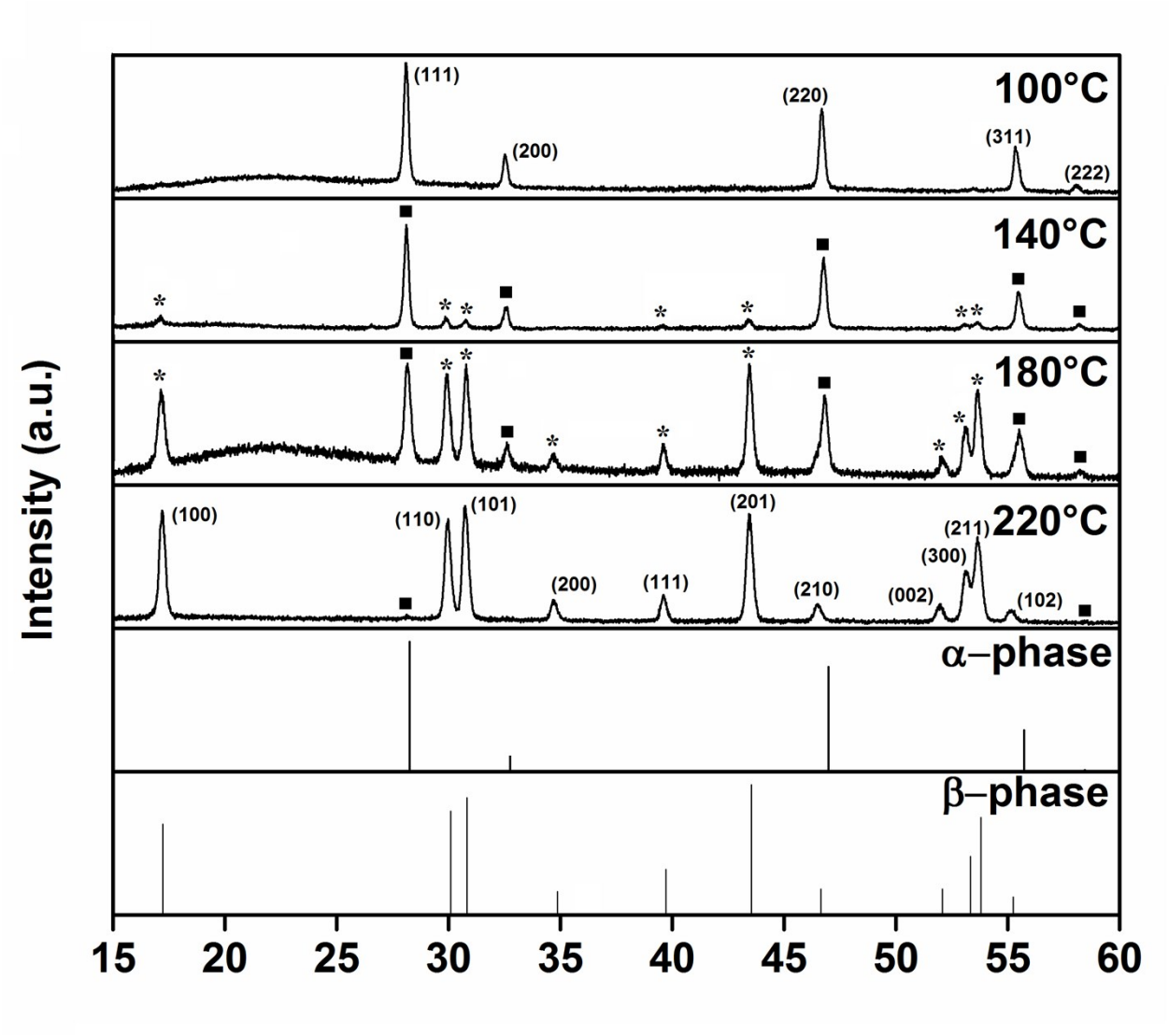

Figure S2. XRD patterns of as-prepared $NaYF_4$ samples, generated with *reaction temperatures* of 100°C, 140°C, 180°C, and 220°C, respectively. The 'stars' designate hexagonal-related facets, while the 'squares' correlate with cubic-related facets. The reaction time has been fixed at 2 h, while the ammonia concentration has been set at 0.2 M, for all four samples processed herein.

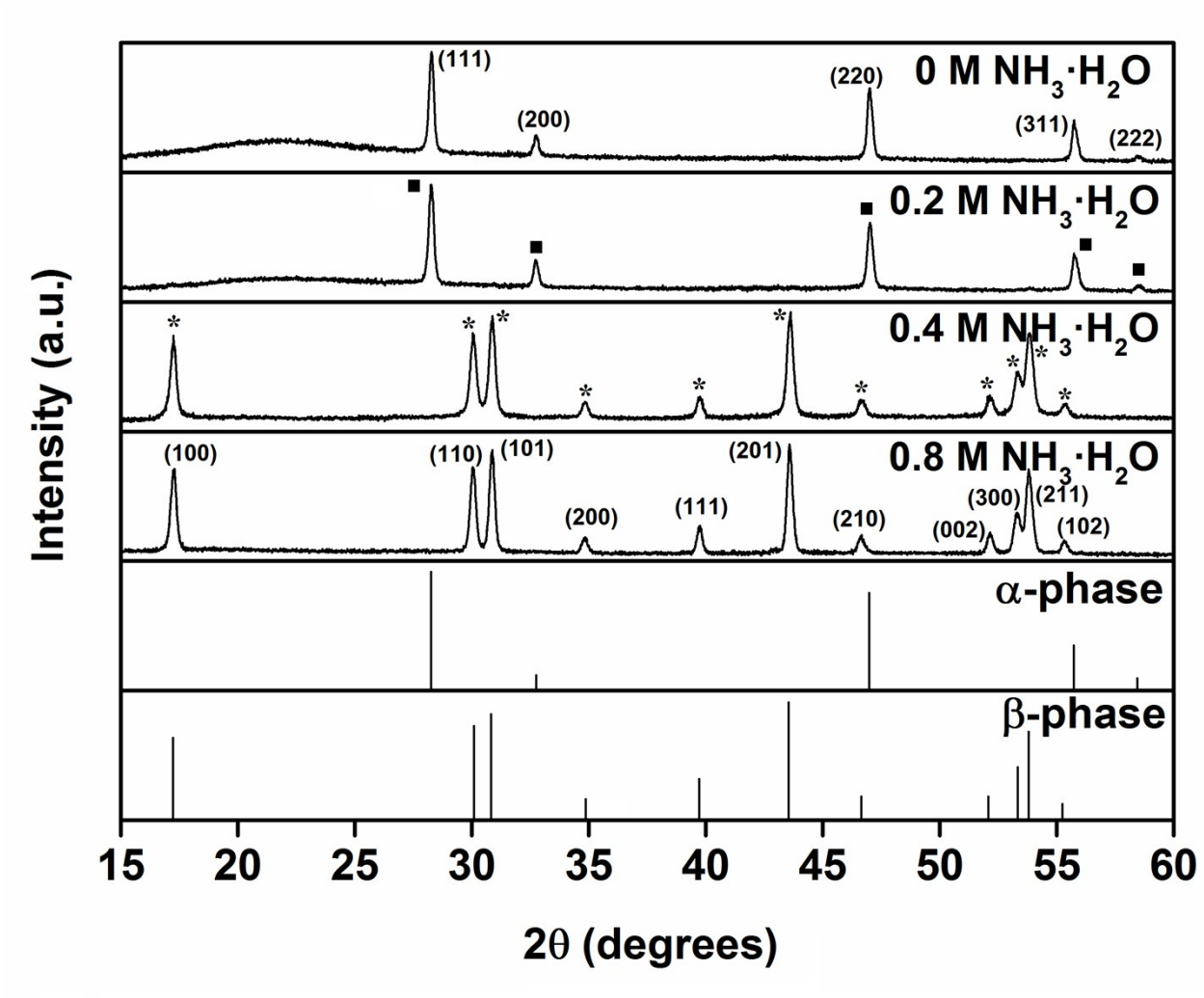

Figure S3. XRD patterns of as-prepared $NaYF_4$ samples, produced with *ammonia concentrations* of 0 M, 0.2 M, 0.4 M, and 0.8 M, respectively. The 'stars' designate hexagonal-related facets, while the 'squares' correlate with cubic-related facets. The reaction time has been fixed at 2 h, while the reaction temperature has been set at 100°C, for all four samples processed herein.

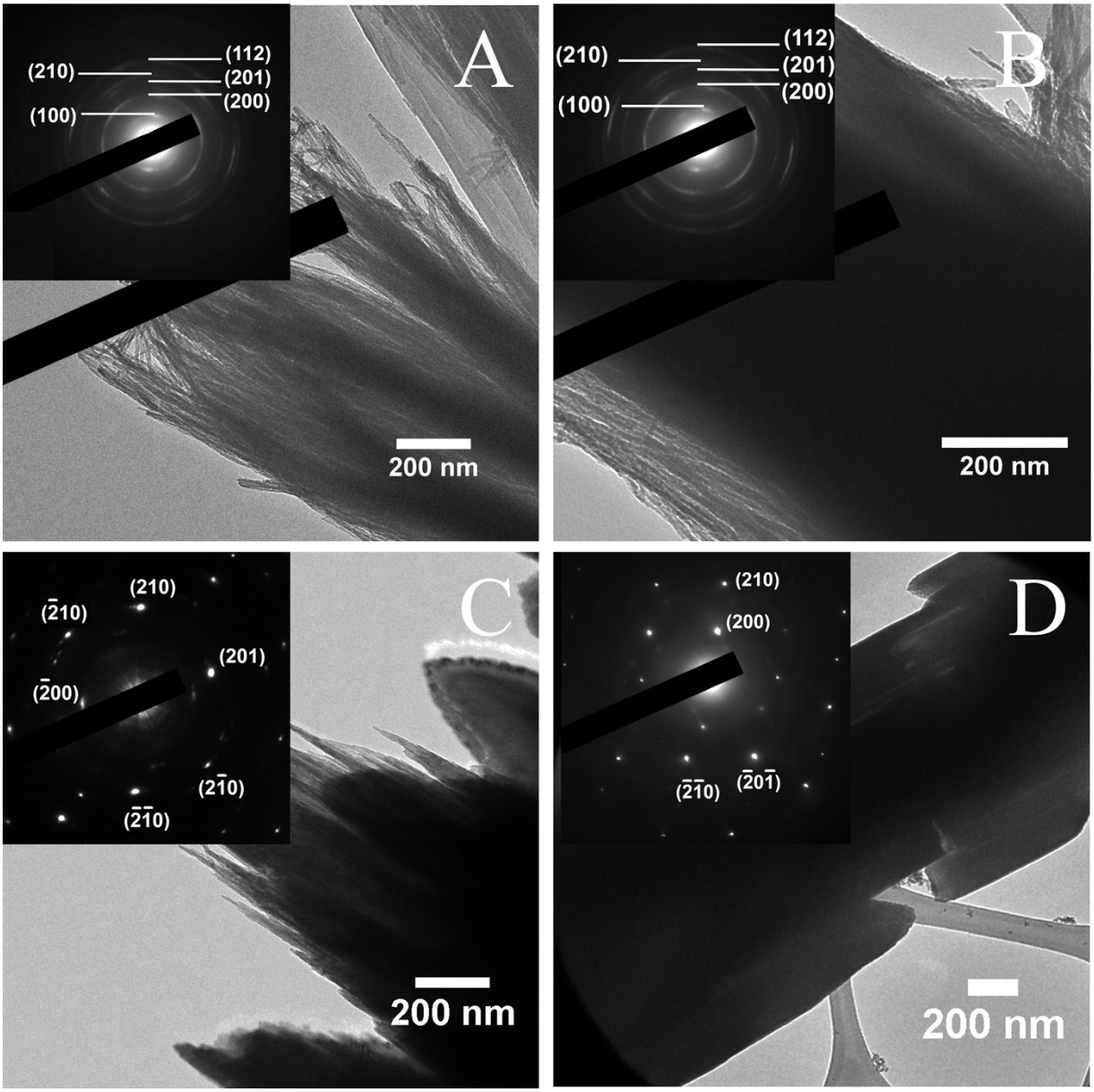

Figure S4. Representative TEM images are shown, focused on the (A) 'edge' and (B) 'central' areas of the **nanowire bundles**. Analogous TEM images are presented, focused on the (C) 'edge' and (D) 'central' regions of **chromosomal-shaped structures**. The insets to (A) through (D) denote typical SAED patterns of these respective regions associated with the two different samples.

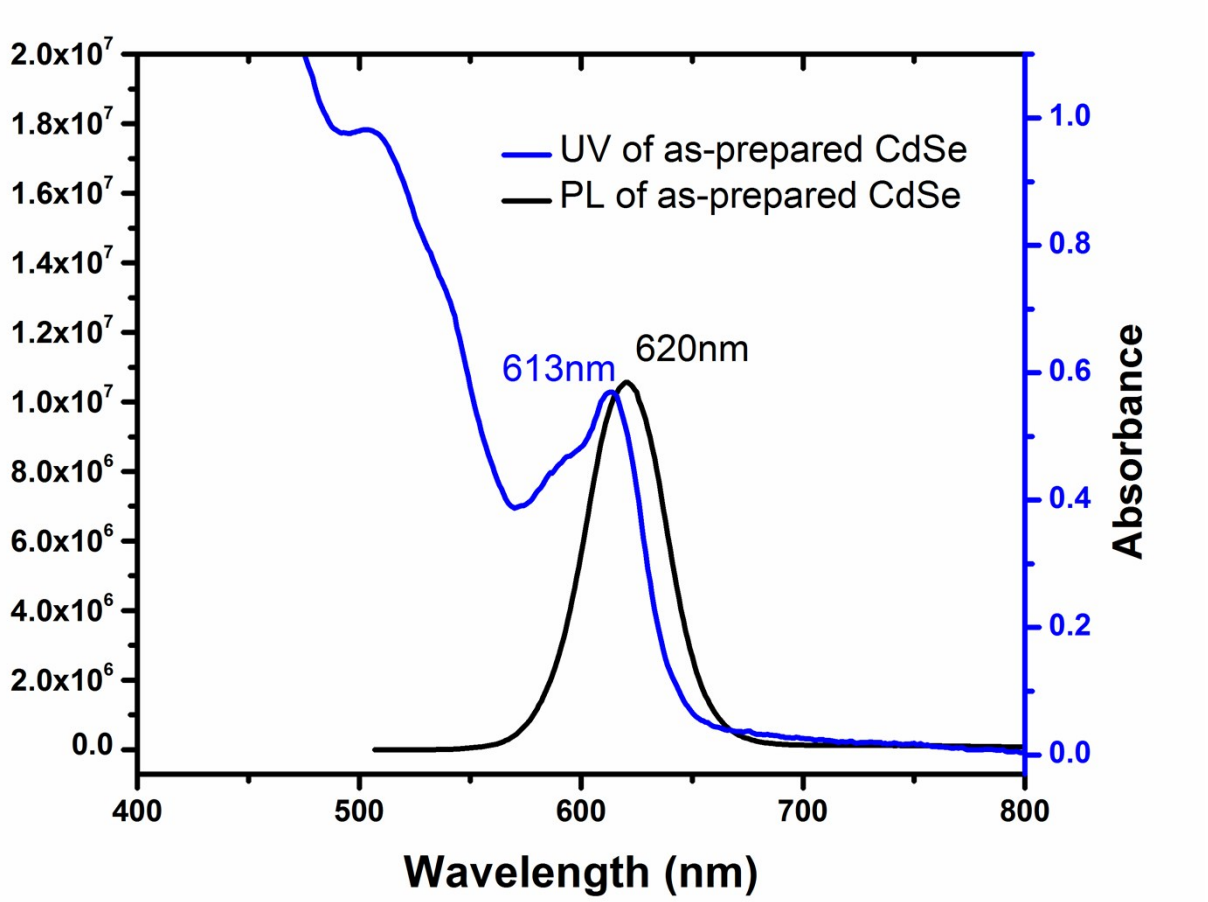

Figure S5. UV-visible and corresponding photoluminescent spectra of as-synthesized CdSe quantum dots. The calculated average size of the QDs is 5.1 nm.

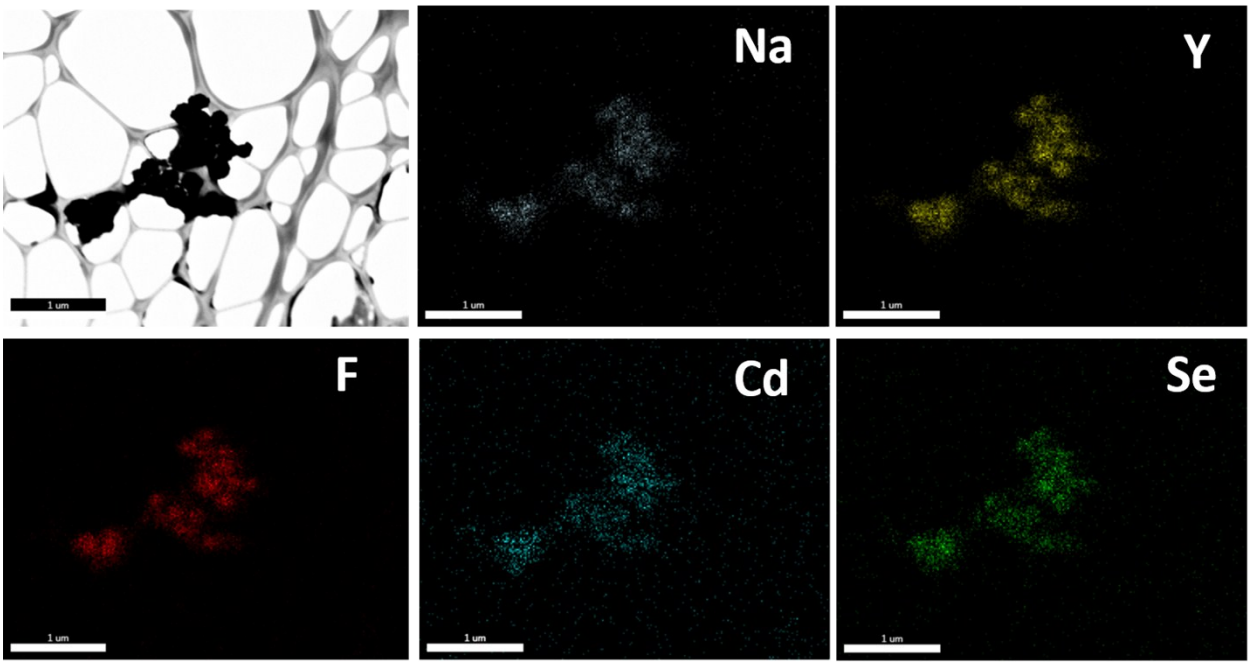

Figure S6. Representative elemental mapping images of 0D-0D $NaYF_4$-CdSe QD heterostructures, created using a nanoparticulate $NaYF_4$ sample (synthesized with 0.2 M ammonia), coupled to as-prepared CdSe quantum dots.

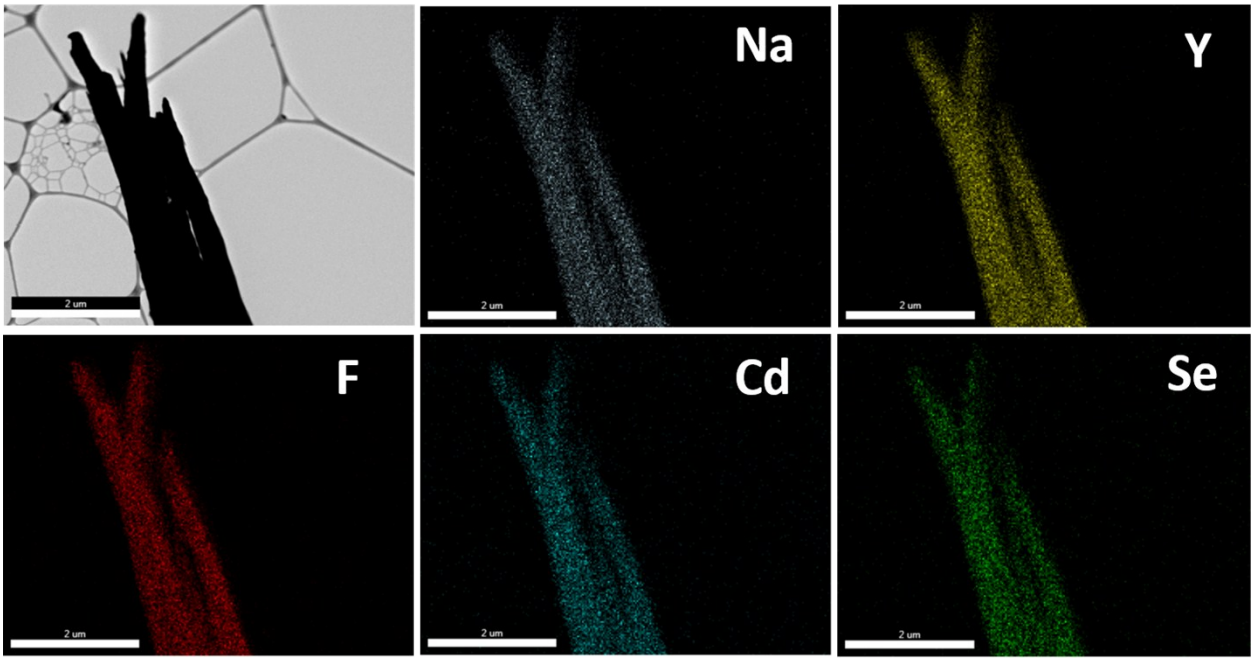

Figure S7. Representative elemental mapping images of 3D-0D $NaYF_4$-CdSe QD heterostructures, created from "nanowire bundles" of $NaYF_4$ (that had been synthesized with 0.8 M ammonia) coupled with as-prepared CdSe quantum dots.

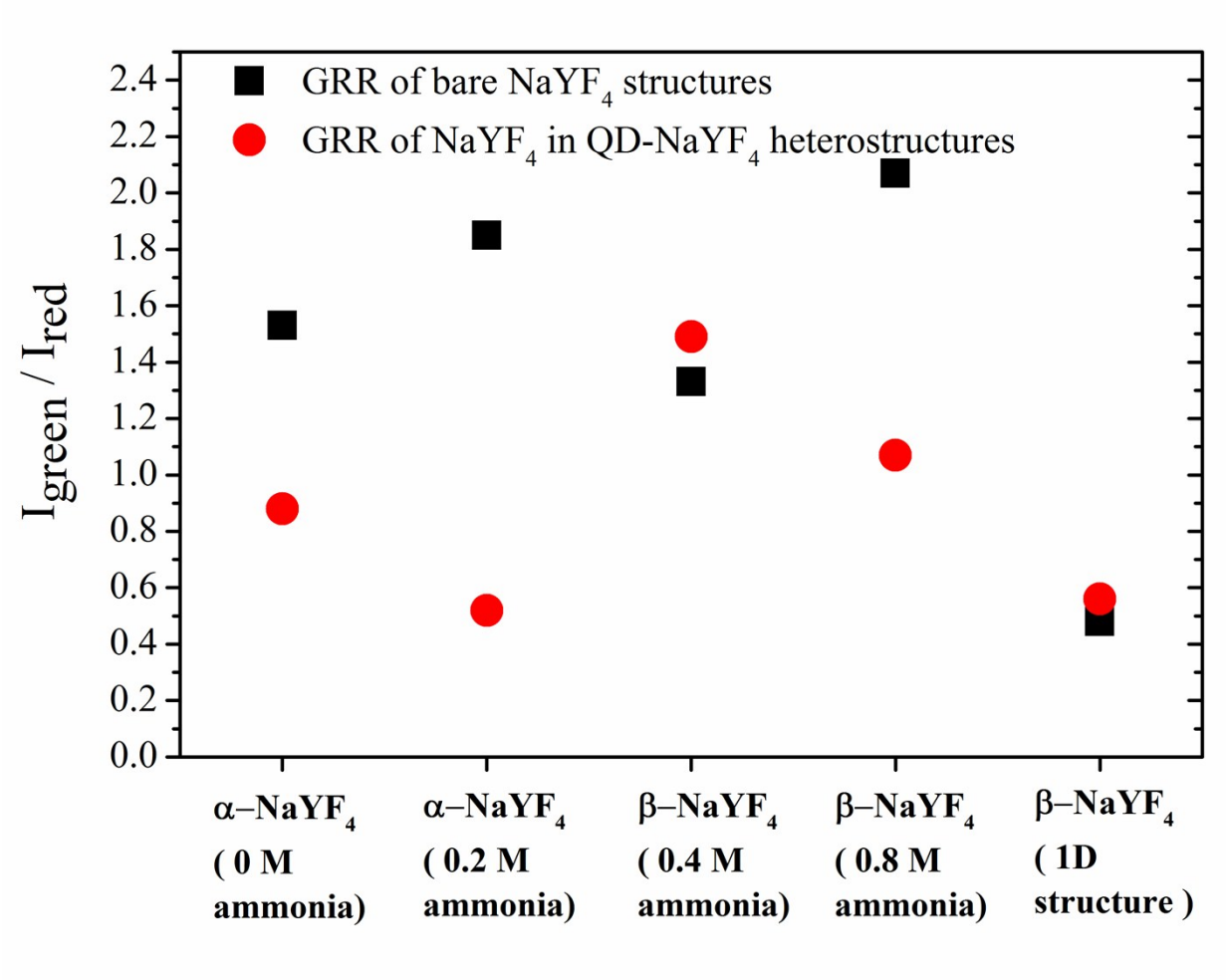

Figure S8. Ratios of green emission to red emission (GRR) intensities of Yb^{3+}, Er^{3+} co-doped $NaYF_4$ samples both before (black rectangles) and after (red dots) CdSe QD attachment onto NaYF4 (including both α and β phases), as a function of the different sample preparative methods described in this paper.

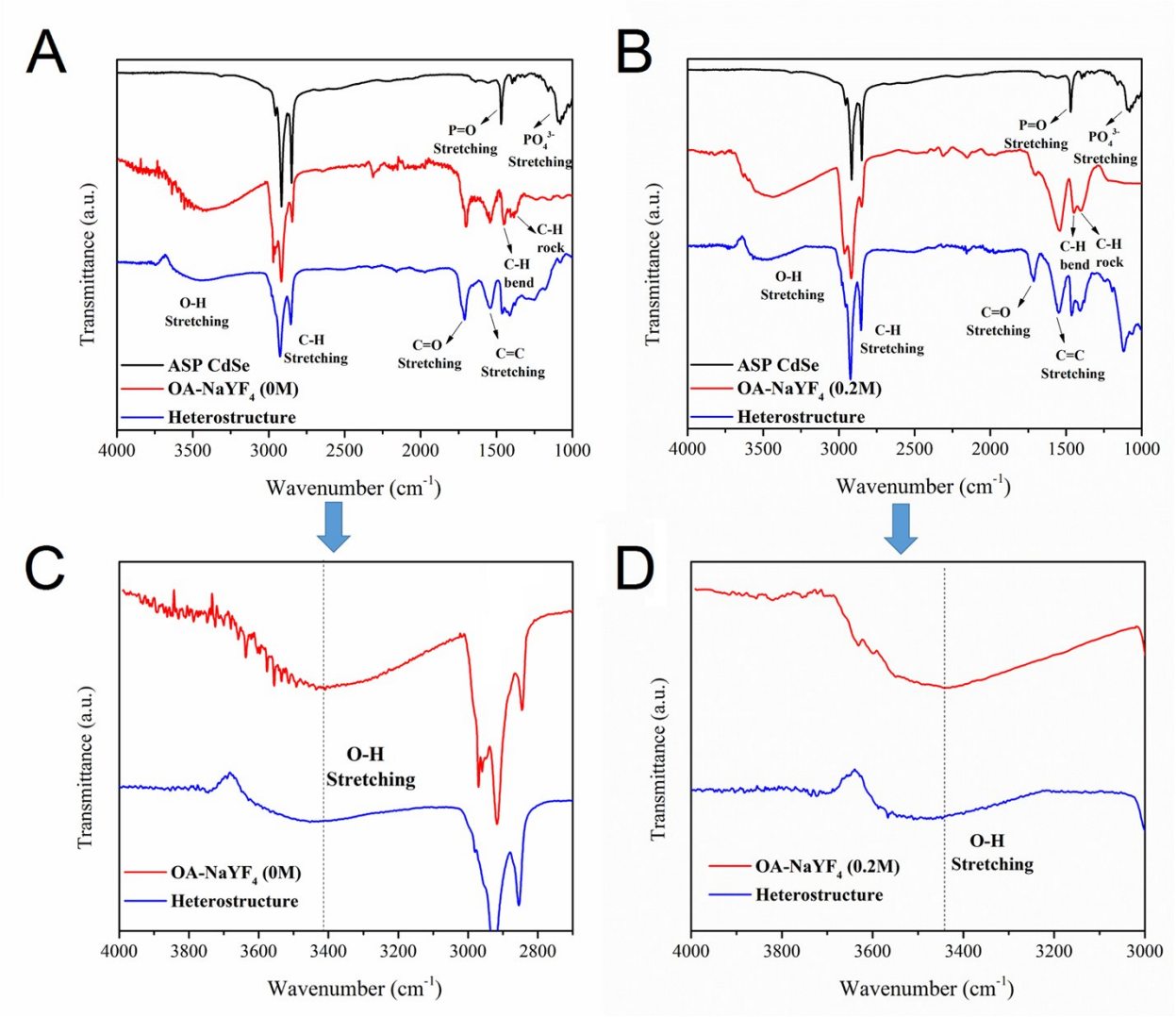

Figure S9. Representative IR spectra of as-prepared CdSe QDs, oleic acid-capped $NaYF_4$ nanoparticles, and the corresponding CdSe QD-$NaYF_4$ heterostructures (A and B). The $NaYF_4$ samples in panel A and B were prepared with 0 M and 0.2 M ammonia, respectively. A magnified view is presented in the range of the O-H stretching mode region (C and D).

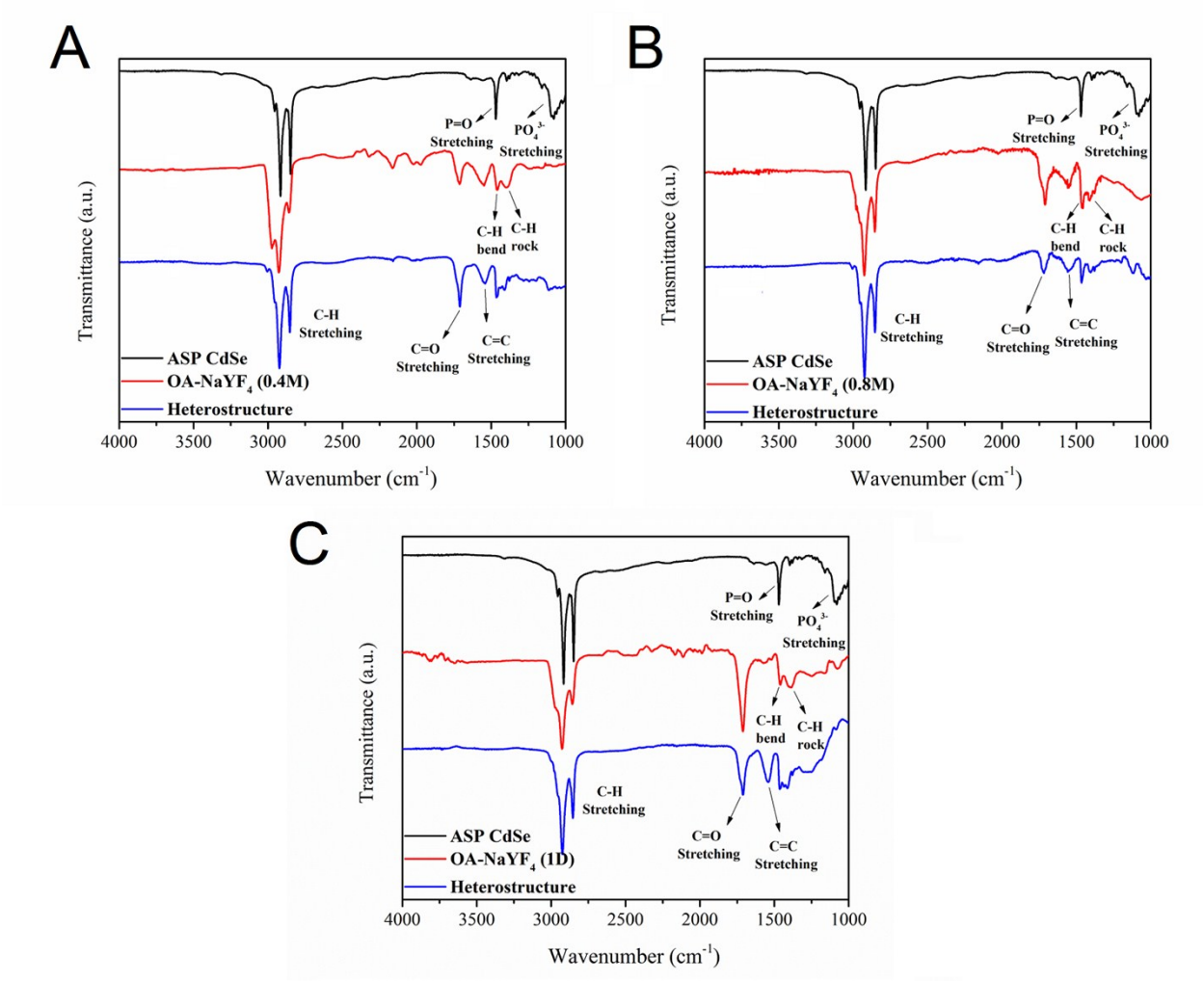

Figure S10. Representative IR spectra of as-prepared CdSe QDs, oleic acid-capped nanoscale motifs of $NaYF_4$ including both (A and B) 3D and (C) 1D morphologies, and the corresponding CdSe QD - $NaYF_4$ heterostructures. The $NaYF_4$ samples in panel A and B were prepared with 0.4 M and 0.8 M ammonia, respectively.