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One-Step Synthesis of White-Light-Emitting Quantum Dots at Low Temperature

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Alloyed $Zn_xCd_{1-x}Se$ quantum dots (QDs) have been successfully prepared at low temperatures by reacting a mixture of $Cd(ClO_4)_2$ and $Zn(ClO_4)_2$ with NaHSe using 3-mercaptopropionic acid as a surface-stabilizing agent. The optical properties and composition of the alloyed QDs were highly dependent on the molar ratio of Zn^{2+} to Cd^{2+}. With an increase in the Zn content, a systematic blue shift in the first exciton absorption and band-edge emission indicated the formation of the alloyed QDs. Moreover, X-ray diffraction peaks of the alloyed QDs systematically shifted to larger angles as the Zn molar fraction of the $Zn_xCd_{1-x}Se$ QDs was increased. This systematic shift further confirmed the appearance of alloyed QDs. Interestingly, among these alloyed QDs, the $Zn_{0.93}Cd_{0.07}Se$ QDs exhibited white-light emission with quantum yields of 12%. In addition, we discovered that we could adjust the $Zn_{0.93}Cd_{0.07}Se$ QD intensity ratio of the band-edge (431 nm) to trap-state (499 nm) emissions by controlling the reaction time. Careful control of the reaction time allowed us to balance the relative strength of the band-edge and trap-state emissions, thereby attaining white-light-emitting QDs. The $Zn_{0.93}Cd_{0.07}Se$ QDs offer unique advantages, including one-step synthesis, tunable white-light emission, easy manipulation, a low-temperature requirement, and low fabrication costs.

Introduction

Semiconductor quantum dots (QDs) have attracted considerable attention not only because of their unique optical and electronic properties[1,2] but also because of their dimensional similarities with biological macromolecules.[3] Additionally, QDs show significant potential in the development of electronic and optical devices such as photodetectors,[4] photovoltaic cells,[5] electronic memory devices,[6] and multicolor light-emitting diodes (LEDs).[7] Because CdSe QDs emit light in the visible range from 500 to 700 nm, they are undoubtedly among the most promising materials used in the fabrication of LEDs. For example, LEDs of various colors have been obtained through a layer-by-layer assembly of charged CdSe QDs and their corresponding oppositely charged polymers.[8,9]

Recently, there has been increasing interest in the development of white-light-emitting LEDs based on QDs. They are expected to have an impact on the lighting industry. Results from previous studies on white-light-emitting organic LEDs have led to the idea that white light could be generated by combining lights of the three primary colors (blue, green, and red).[10,11] However, most of the white-light-emitting organic LEDs suffer from complicated doping schemes and combinations of several organic components.[10,11] Unfortunately, a similar phenomenon is encountered in the development of white-light-emitting LEDs based on QDs.[12,13] Although QDs require only a single excitation source for emitting lights of multiple colors, the weight ratio among the red-, green-, and blue-emitting QDs should be elaborately adjusted to obtain white-light emission. Additionally, the fluorescence resonance energy transfer between QDs should be considered when the emission spectrum of blue-emitting QDs overlaps the excitation spectrum of green/red-emitting QDs.[14] On the basis of the above-mentioned results, we

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suggest that single-emitting components can offer many advantages over multiple-emitting components for white-light-emitting LEDs, including better reproducibility, low-cost preparation, easy modification, and simple fabrication processes. However, few researches have been conducted on the synthesis of white-light-emitting QDs.[15-18] Using organic surfactants such as capping agents, ultrasmall CdSe QDs synthesized at high or room temperature had narrow distributions in size but exhibited a broad-band emission (420−710 nm).[15] These kinds of QDs were capable of emitting white light, and their quantum yield was 2−3%. When oleic acid was used instead of organic surfactants, the quantum yield of white-light-emitting CdS QDs was enhanced up to 17%.[16] Moreover, the onionlike CdSe/ZnS/CdSe/ZnS QDs displayed white-light emission with a quantum yield of 30%, which was obtained by adjusting the relative emission strengths between the orange-emitting (CdSe) and blue-emitting (ZnS) components.[16]

Herein, we report a new approach for the one-step synthesis of white-light-emitting alloyed QDs (ternary $Zn_xCd_{1-x}Se$) stabilized with 3-mercaptopropionic acid (MPA) in the aqueous phase. It should be noted that the aim of this study is to develop a simple, reliable, and inexpensive method for the synthesis of white-light-emitting QDs with high quantum yield. The composition and optical properties of the alloyed QDs were determined by adjusting the molar ratio of Zn^{2+} to Cd^{2+} (the precursor solution) and controlling the reaction conditions (temperature and time). These factors strongly influenced the emission wavelength, the emission intensity, and the intensity ratio of the band-edge to trap-state emissions. By adjusting the relative strength of the band-edge and trap-state emission, we unveiled that the $Zn_{0.93}Cd_{0.07}Se$ QDs were capable of generating broad-band emission (376−720 nm) throughout most of the visible light spectrum, resulting in white-light emission. The obtained $Zn_{0.93}Cd_{0.07}Se$ QDs (single-emitting components) will be promising nanomaterials that can be used in the manufacture of white-light-emitting LEDs.

Experimental Section

Materials. Cadmium perchlorate, zinc perchlorate, 3-mercaptopropionic acid (MPA), sodium borohydride, selenium powder, sodium hydroxide, 2-propanol, and Rhodamine 6G were purchased from Aldrich (Milwaukee, WI). The commercial LEDs were obtained from Everlight (334-15/W2C1 series, Taipei, Taiwan) and driven by a 2.7 V constant-voltage source. Milli-Q ultrapure water was used in all of the experiments.

Preparation of NaHSe. The synthesis of alloyed QDs was based on the reaction of $Zn(ClO_4)_2$ and $Cd(ClO_4)_2$ with NaHSe. All reactions were performed in oxygen-free water under nitrogen. Sodium borohydride (0.08 g) was dissolved in 1 mL of ultrapure water. Sodium hydroselenide was prepared by mixing sodium borohydride (1.0 mL) and selenium powder (0.08 g). Approximately 8 h later, the black selenium powder was reduced completely and a dark-red solution of NaHSe was formed.

Synthesis of Alloyed QDs. Before the addition of NaHSe, the pH of the solution containing MPA (11.5 M, 26.4 μL), $Zn(ClO_4)_2$ (0.1 M, 612−1212 μL), and $Cd(ClO_4)_2$ (0.1 M, 12.1−612 μL) was adjusted to 11.0 with 1.0 M NaOH. The total volume of the solution was added up to 15 mL with ultrapure water. Subsequently, 20 μL of a freshly prepared NaHSe solution was injected into the resulting solution. The molar ratios (volume) of Zn^{2+}/Cd^{2+} were 1:1 (612 μL:612 μL), 10:1 (1112.7 μL:111.3 μL), 35:1 (1190 μL:34.0 μL), and 100:1 (1212 μL:12.1 μL). Under these conditions, the total amount of $Zn(ClO_4)_2$ and $Cd(ClO_4)_2$ was a constant 0.1224 mmol, about 6-fold excess of NaHSe in these solutions. Finally, the resulting mixture was reacted at 80 °C for 20 h to promote the growth of the alloyed QDs and then rapidly cooled to room temperature. No aggregation and no significant changes in their spectral characteristics were detected over several months of being stored at 4 °C in the dark.

Synthesis of CdSe and ZnSe QDs. A total of 20 μL of a freshly prepared NaHSe solution was added to a solution (15 mL) containing MPA (11.5 M, 26.4 μL) and $Zn(ClO_4)_2$ (0.1 M, 1224 μL) at pH 11.0 with vigorous stirring. The resulting mixture was heated at 80 °C for 20 h. In the case of the preparation of CdSe QDs, $Cd(ClO_4)_2$ was used in place of $Zn(ClO_4)_2$ in the precursor solution.

Characterization of QDs. A Cintra 10e double-beam ultraviolet−visible (UV−vis) spectrophotometer (GBC, Victoria, Australia) was used to examine the absorption spectra of the QDs. An Aminco−Bowman fluorometer (ThermoSpectronic, Pittsford, NY) was used to measure the fluorescence spectra of the QDs while irradiating their solutions at 340 nm. The average sizes of white-light-emitting QDs were estimated by conducting high-resolution transmission electron microscopy (HRTEM) measurements (Tecnai G2 F20 S-Twin). Energy-dispersive X-ray spectra (EDS) were obtained using a HRTEM microscope. Powder X-ray diffraction (XRD) patterns were obtained using a diffractometer (model X'Pert Pro; PANalytical, Spectris plc, Almelo, The Netherlands) with Cu Kα radiation (λ = 1.5418 Å). The quantum yields of the QDs were estimated from the integrated fluorescence intensities of the QDs and the reference (Rhodamine 6G in ethanol at room temperature, quantum yield = 95%). Photoimages of the QD solutions, which were illuminated using a UVGL-25 UV lamp (365 nm; Upland, CA), were colloected using a Coolpix 5400 digital color camera (Nikon, Tokyo, Japan).

To purify the alloyed QDs, they were precipitated with 2-propanol according to the previous studies.[19] First, 2-propanol was added dropwise under stirring until the QD solution became slightly turbid. The resulting solution was subjected to two centrifuge−wash cycles to remove the excess Zn^{2+}, Cd^{2+}, NaHSe, MPA, and solutes that were physically adsorbed onto the surface of the as-prepared QDs. The centrifugation was conducted at 3000 rpm for 10 min, and 2-propanol was used for washing in each cycle. The purified QDs were then digested in a standard HCl/HNO_3 solution for inductively coupled plasma mass spectrometry (ICP-MS) analyses that were performed using ELAN 6100 DRC ICP-MS measurement (Perkin-Elmer-SCIEX, Thornhill, Ontario, Canada). The concentrations of Zn and Cd ions in the digested solution were estimated from the calibration curves of the standard solutions. As a result, we can determine the composition of the alloyed $Zn_xCd_{1-x}Se$ QDs.

Solid-State Lighting Device. Poly(dimethylsiloxane) (PDMS; Sylgard 184, Dow Corning, Midland, MI) was prepared by blending a base and a curing agent in a mixing ratio of 10:1. The white-light-emitting $Zn_{0.93}Cd_{0.07}Se$ QDs (10×; 1 mL) were added to the liquid-gel-type mixture (9 mL). It is to be noted that the 1× $Zn_{0.93}Cd_{0.07}Se$ QDs were obtained from the reaction of 8 mM Zn^{2+}, 0.23 mM Cd^{2+}, and 1.3 mM NaHSe in the

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Table 1. Optical Properties of the $Zn_xCd_{1-x}Se$ QDs

molar ratio (Zn/Cd)	composition (x in $Zn_xCd_{1-x}Se$)	first exciton absorption (nm)	band-edge emission (nm)[a]	trap-state emission (nm)[a]	size (nm)[b]	quantum yield (%)
1:1	0.02	540	564		2.2	0.1
10:1	0.30	467	473	550	2.5	7
35:1	0.93	418	431	499	2.7	12
100:1	0.97	403	416	482	2.3	15

[a] Excited at 340 nm. [b] The size was estimated from the XRD pattern.

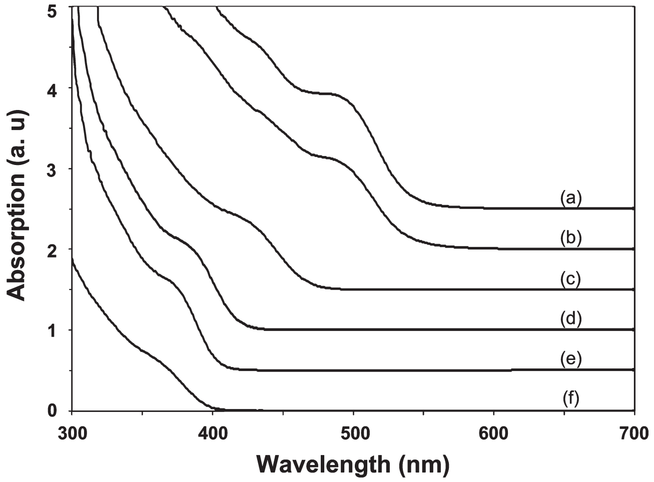

Figure 1. Absorption spectra for the $Zn_xCd_{1-x}Se$ QDs with Zn mole fractions of (a) 0.00, (b) 0.02, (c) 0.30, (d) 0.93, (e) 0.97, and (f) 1.00.

presence of MPA. The resulting mixture was stirred at room temperature until the solution became homogeneous. After that, homogeneous solutions were degassed in a vacuum chamber. A glass slide was coated with the resulting mixture and left in the chamber for 24 h at room temperature. The 365-nm UV lamp was used as the pump source. The device consisted of a UV lamp and a glass slide coated with a thin film of alloyed QDs.

Results and Discussion

Effect of the Zn^{2+}/Cd^{2+} Molar Ratio. Previous studies have reported that the composition of the $CdSe_{1-x}Te_x$ QDs is highly dependent on the Se/Te molar ratio in the precursor solution and on the intrinsic reactivities of Se and Te toward Cd^{2+}.[20] On the basis of this result, we assumed that the composition of the $Zn_xCd_{1-x}Se$ QDs was strongly affected by the Zn^{2+}/Cd^{2+} molar ratio in the precursor solution and by the reactivities of Zn^{2+} and Cd^{2+} toward NaHSe. Although their intrinsic reactivities are difficult to change, the Zn^{2+}/Cd^{2+} molar ratio in the precursor solution provides a convenient control parameter for the preparation of composition-tunable $Zn_xCd_{1-x}Se$ QDs. First, we used ICP-MS to determine the composition of the alloyed QDs. In Table 1, the prepared Zn^{2+}/Cd^{2+} molar ratios of 1:1, 10:1, 35:1, and 100:1 resulted in nanocrystal compositions of 2:98, 30:70, 93:7, and 97:3, respectively. Note that the reaction temperature and time for the synthesis of the QDs were fixed at 80 °C and 20 h, respectively. Figure 1 shows the absorption spectra of a series of ZnSe, $Zn_xCd_{1-x}Se$, and CdSe QDs. The bulk ZnSe and CdSe composites have band gaps of 2.67 and 1.74 eV, which correspond to the absorption band edges of 460 and 712 nm, respectively.[21] Compared with the absorption spectrum of the CdSe QDs (spectrum a in Figure 1), the $Zn_{0.02}Cd_{0.98}Se$ QDs presented a similar absorption feature (spectrum b in Figure 1). When the Zn molar fraction in the $Zn_xCd_{1-x}Se$ QDs was increased from 0.02 to 0.97, a systematic blue shift for the first exciton absorption peak of the $Zn_xCd_{1-x}Se$ QDs was observed (spectra b−e in Figure 1 and Table 1). The first exciton absorption peak of each $Zn_xCd_{1-x}Se$ QD was obviously located between those of the ZnSe and CdSe QDs. Moreover, the absorption spectrum of a mixture of ZnSe and CdSe QDs displayed two absorption features at approximately 406 and 530 nm (Figure S1 in the Supporting Information), which was evidently different from the absorption spectrum of each $Zn_xCd_{1-x}Se$ QD with a single feature. Therefore, the separate formation of CdSe and ZnSe QDs during the reaction can be ruled out. Moreover, the first exciton absorption peak of each $Zn_xCd_{1-x}Se$ QD is not as clear as that of CdSe QD. These poorly resolved absorption spectra are characteristic of the $Zn_xCd_{1-x}Se$ QDs. Similar phenomena have been reported for the synthesis of the $Zn_xCd_{1-x}Se$ QDs.[22,23] On the basis of our results and the published data, it is suggested that a broad and poorly resolved absorption band is not caused by the wide size distribution. The photoluminescence (PL) spectra of a series of ZnSe, $Zn_xCd_{1-x}Se$, and CdSe QDs are shown in Figure 2. As expected, the PL spectrum of the CdSe QDs resembled that of the $Zn_{0.02}Cd_{0.98}Se$ QDs (spectra a and b in Figure 2) because they had similar absorption spectra. When the Zn molar fraction in the $Zn_xCd_{1-x}Se$ QDs was increased to 0.30, the $Zn_{0.3}Cd_{0.7}Se$ QDs thus obtained exhibited a narrow emission peak at 473 nm and a broad peak at 550 nm (spectrum c in Figure 2). A small Stokes shift (~6 nm) between the first exciton absorption peak (spectrum c in Figure 1) and the narrow emission peak (spectrum c in Figure 2) indicates that the peak at 473 nm originated from the band-edge emission.[24] In contrast, a large red shift was observed during a comparison of the first exciton absorption peak and the broad peak. The peak at 550 nm may be related to the trap-state emission, which was induced by surface defects (hole-trapping thiolates on the particle surface).[25] Similar phenomena have been previously seen in the case of syntheses of

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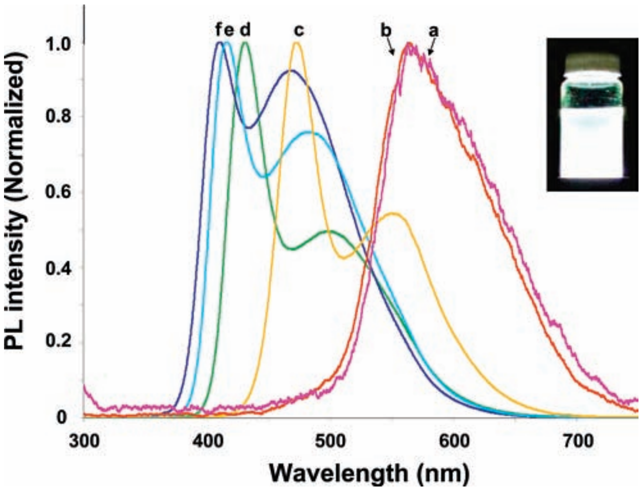

Figure 2. PL spectra for the $Zn_xCd_{1-x}Se$ QDs with Zn mole fractions of (a) 0.00, (b) 0.02, (c) 0.30, (d) 0.93, (e) 0.97, and (f) 1.00. The excitation wavelength was set at 340 nm. Inset: Fluorescent photoimage of the $Zn_{0.93}Cd_{0.07}Se$ QDs excited by a 365-nm UV lamp.

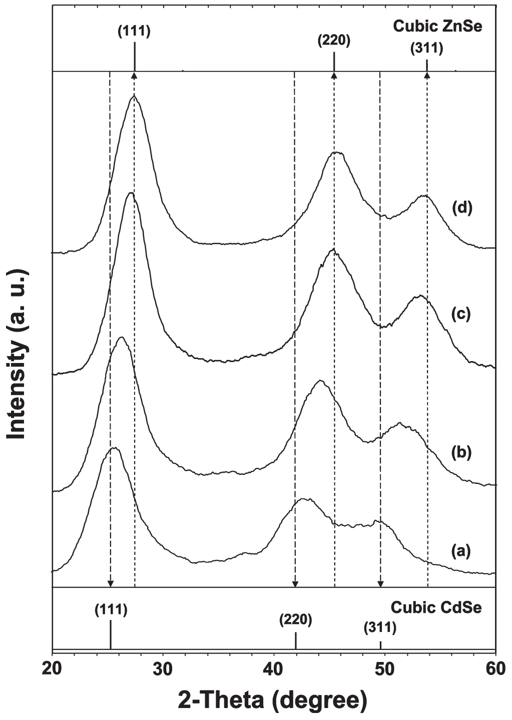

Figure 3. Powder XRD patterns of the $Zn_xCd_{1-x}Se$ QDs with Zn mole fractions of (a) 0.02, (b) 0.30, (c) 0.93, and (d) 0.97.

MPA-capped ZnSe QDs.[23,25,26] As shown in spectra d−f in Figure 2, the intensity ratio of the band-edge to trap-state emissions was reduced as the Zn molar fraction in the $Zn_xCd_{1-x}Se$ QDs was increased from 0.93 to 1.00. In addition, we observed a systematic blue shift in the band-edge emission peak when the Zn molar fraction was varied from 0.00 to 1.00. This significant blue shift indicates the formation of the alloyed $Zn_xCd_{1-x}Se$ QDs. It was impossible to separate the CdSe and ZnSe QDs in the spectrum because we did not find their corresponding PL spectra. Moreover, if CdSe (core) and ZnSe (shell) QDs were formed, their PL peak should shift to a longer wavelength after the CdSe core has been coated with the ZnSe shell.[27−29] This unique blue shift of the alloyed QDs has also been reported for the syntheses of the $Zn_xCd_{1-x}S$ and $Zn_xCd_{1-x}Se$ QDs.[22,30]

We also used powder XRD to obtain patterns in an effort to verify the alloyed structure. The XRD patterns of bulk cubic CdSe and ZnSe are shown at the bottom and top of Figure 3, respectively. The XRD peaks of the $Zn_xCd_{1-x}Se$ QDs were apparently located between those of the bulk CdSe and ZnSe composites. When the Zn molar fraction increased, the XRD peaks gradually shifted toward larger angles. This shift in the XRD peaks is indicative of the transformation from a CdSe lattice to a ZnSe lattice. The continuous peak shifting of the alloyed QDs supported our tendency to rule out the separate formation of the ZnSe and CdSe QDs. This phenomenon has been commonly observed in other published studies on the syntheses of alloyed QDs.[20,22,23,30−32] Meanwhile, the average crystallite sizes of the $Zn_{0.02}Cd_{0.98}Se$, $Zn_{0.30}Cd_{0.70}Se$, $Zn_{0.93}Cd_{0.07}Se$, and $Zn_{0.97}Cd_{0.30}Se$ QDs were 2.2, 2.5, 2.7, and 2.3 nm, respectively, as calculated from the (111) peak using the Scherrer equation.[23]

Characterization of White-Light-Emitting QDs. We discovered white-light emission from the alloyed $Zn_{0.93}Cd_{0.07}Se$ QDs ranging from 376 to 720 nm in wavelength (inset to Figure 2). The fluorescent spectrum of the $Zn_{0.93}Cd_{0.07}Se$ QDs consists of a band-edge emission at 431 nm and a trap-state emission at 499 nm (spectrum d in Figure 2). A similar spectral pattern had also been observed in white-light-emitting devices.[33−35] The fluorescence quantum yield (12%) of the resulting nanocrystals is comparable with that of the previously reported CdS QDs with white-light emission.[16] When we compared the $Zn_{0.93}Cd_{0.07}Se$ QD emission with the different color temperatures (warm, cold, and pure white) of commercial white-light LEDs, we found that the emission belonged in the cold-white-light classification (Figure S2 in the Supporting Information). The Commission Internationale de l'Eclairage has assigned the following standard chromaticity coordinates to commercial cold-white-LED illumination: $X = 0.264{-}0.296$ and $Y = 0.248{-}0.305$. The composition and size of the $Zn_{0.93}Cd_{0.07}Se$ QDs were examined by EDS and HRTEM. As indicated in Figure 4A, the EDS spectrum reveals the existence of Zn, Se, and Cd in the specimen. The relative Zn/Cd intensity ratio revealed the composition of the as-prepared QDs. The main source of S was from the stabilizer MPA. The HRTEM images illustrated in Figure 4B indicate that the nanocrystals had

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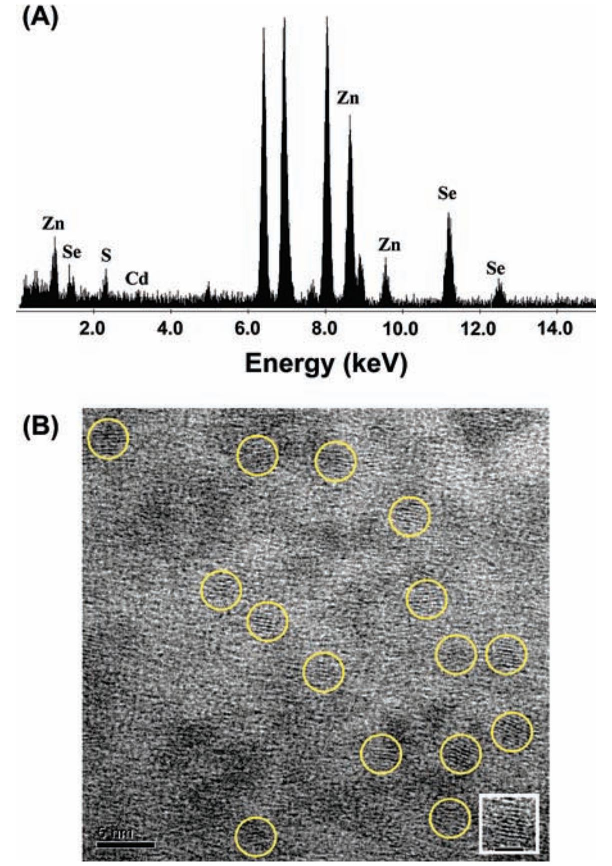

Figure 4. (A) EDS spectrum and (B) HRTEM image of the $Zn_{0.93}Cd_{0.07}Se$ QDs. The yellow circles in part A indicate the $Zn_{0.93}Cd_{0.07}Se$ QDs, and the scale bar in the inset of the HRTEM image was 2.5 nm.

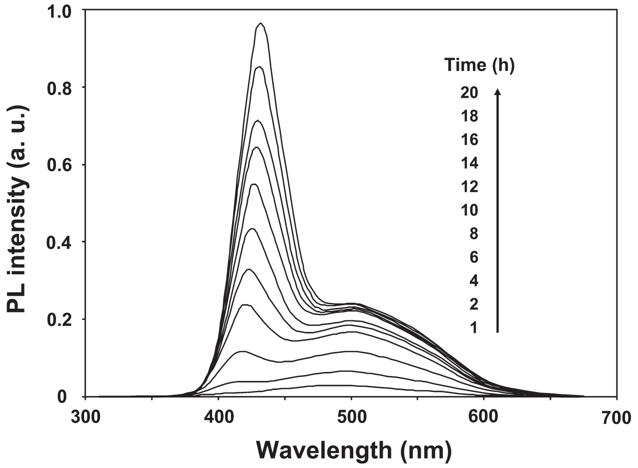

Figure 5. PL spectra obtained from the $Zn_{0.93}Cd_{0.07}Se$ QDs at different growth times.

an average diameter of 2.72 ± 0.46 nm ($n = 60$). The size of the $Zn_{0.93}Cd_{0.07}Se$ QDs obtained by the HRTEM images is consistent with that obtained by XRD.

Growth Mechanism. To understand the formation mechanism of the white-light-emitting $Zn_{0.93}Cd_{0.07}Se$ QDs, we investigated the growth kinetics of white-light-emitting QDs by measuring the PL spectra and by obtaining XRD patterns at different growth stages. As shown in Figure 5, the PL spectra of the QDs were obtained in the time range of 1−20 h. With an increase in the reaction time, both band-edge and trap-state emissions of PL spectra shifted to a longer wavelength. This shift was due to the increase in the particle size.[20] In addition, the PL spectra of the QDs mainly consisted of trap-state emission in the reaction time range of 1−4 h, whereas their spectra consisted of both band-edge and trap-state emissions above 4 h. This finding reveals that the smaller QDs exhibited a higher density of surface defects at the early stage of particle growth.[25] Compared with the trap-state emission, the band-edge emission of the alloyed QDs was enhanced dramatically when the heating time was increased from 1 to 20 h. The band-edge emission was probably enhanced because the diffusion of the inner atoms increased with the reaction time. As a result, the composition of the QDs obtained at the late stage of particle growth was more homogeneous than that obtained at the early stage. To support this hypothesis, the prepared Zn^{2+}/Cd^{2+} molar ratio of 35:1 was synthesized at a higher temperature (120 °C) in the presence of NaHSe and MPA. Note that diffusion of the inner atoms was more efficient at higher temperatures. Figure S3 (Supporting Information) illustrates that the alloyed QDs consisted of a strong band-edge emission and a weak trap-state emission at a higher reaction temperature. This result provides clear evidence that the improvement in the compositional homogeneity of alloyed QDs can efficiently enhance band-edge emission and suppress trap-state emission (removal of surface defects).[30] On the basis of these results, we suggest that the enhancement in the band-edge emission of the alloyed QDs found in our study was mainly attributable to the improvement of their compositional homogeneity rather than to the change in their compositional fraction. In addition, we examined the XRD pattern of the alloyed QDs at different growth times (Figure 6). The XRD peak pattern remained almost constant as the reaction time increased, implying that the composition ratio of the alloyed QDs was almost unchanged when the reaction time was increased from 1 to 20 h. To further support the formation of the alloyed QDs, ICP-MS was used for elemental analysis at different growth intervals. Figure S5 (Supporting Information) reveals that only a slight decrease of ca. 2% in the Cd content was observed from the beginning (1 h) to the end of particle growth. This result indicates a nearly constant composition for the QDs during the entire course of their growth. Fluorescent photoimages of the white-light-emitting $Zn_{0.93}Cd_{0.07}Se$ QDs at different growth times are shown in Figure S4 (Supporting Information). White-light emission was observed at intensity ratios of the band-edge to trap-state emissions between 2.0 and 3.0 exhibited. Thus, by carefully controlling the reaction time, we were able to adjust the relative strength of the band-edge and trap-state emissions, thereby obtaining white-light emission. However, it is impossible for white-light-emitting CdSe or CdS QDs to perform this type of emission tuning.[15,16] In addition, although the onionlike CdSe/ZnS/CdSe/ZnS QDs exhibited high quantum yield, multistep synthesis was required to obtain white-light emission.[16] Consequently, the synthesis of $Zn_{0.93}Cd_{0.07}Se$ QDs offers multiple advantages, including one-step synthesis, tunable white-light emission, easy manipulation, low-temperature requirements, and low fabrication costs. To the best

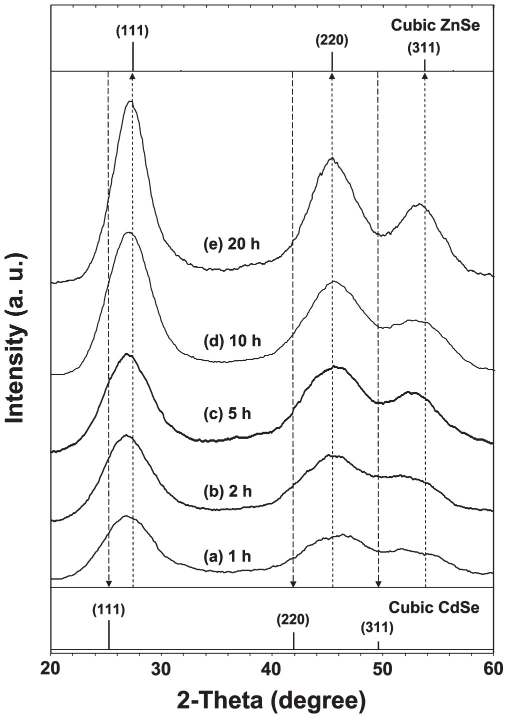

Figure 6. Powder XRD patterns of the $Zn_{0.93}Cd_{0.07}Se$ QDs at different growth times: (a) 1, (b) 2, (c) 5, (d) 10, and (e) 20 h.

of our knowledge, this is the first example where the alloyed $Zn_xCd_{1-x}Se$ QDs are capable of emitting white light.

Nanocrystals as Phosphor. The successful synthesis of the alloyed $Zn_{0.93}Cd_{0.07}Se$ QDs, as described earlier, motivated us to apply the synthesized QDs to the development of solid-state light sources for general illumination. Typically, the use of semiconductor nanocrystals can provide important advantages over conventional phosphors, such as size-tunable band gaps, high-fluorescence quantum efficiencies, good photostability, low fabrication costs, and chemical flexibility.[36] In addition to the advantages mentioned above, the $Zn_{0.93}Cd_{0.07}Se$ QDs alleviate the complications associated with the design and fabrication of white-light-emitting, solid-state devices because their broad-band radiation was used as a single-emitting component. High concentrations of an organic dye can cause fluorescence self-quenching, whereby the intensity of the fluorescence is decreased as the dye molecules collide with one another. This phenomenon was also observed in high concentrations of QDs. Figure 7A shows how the concentration of the QDs affects their PL intensity. This indicates that the PL intensity began to decline when the concentration of the $Zn_{0.93}Cd_{0.07}Se$ QDs exceeded 1×, which was prepared in the precursor mixture of 7.9 mM Zn^{2+}, 0.2 mM Cd^{2+}, and 1.3 mM NaHSe in the presence of MPA. We fabricated a white-light illumination source that contained a single film of 1× alloyed QDs blended with UV-transparent PDMS. The thin $Zn_{0.93}Cd_{0.07}Se$ QD film (coated on a glass slide) maintained its optical properties even after 10 days of exposure to a commercial UV lamp (365 nm, 70 mW/cm^2) under ambient conditions. Figure 7B displays a photo of a white-light-emitting device in operation.

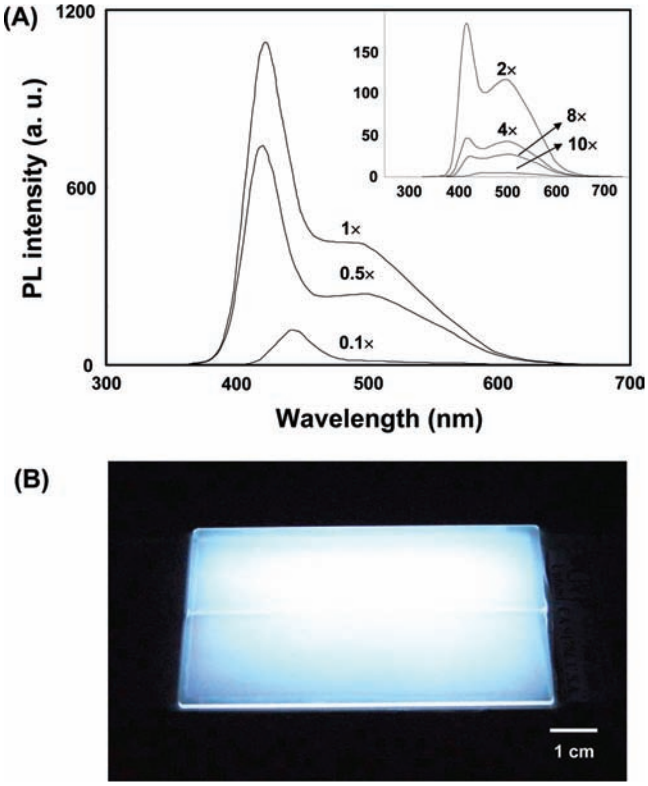

Figure 7. (A) Effect of the concentration of the $Zn_{0.93}Cd_{0.07}Se$ QDs on the PL intensity. The 1× $Zn_{0.93}Cd_{0.07}Se$ QDs were obtained from the reaction of 8 mM Zn^{2+}, 0.23 mM Cd^{2+}, and 1.3 mM NaHSe in the presence of MPA. (B) Thin coating of the $Zn_{0.93}Cd_{0.07}Se$ QDs on a glass slide illuminated by a commercial UV lamp.

Conclusions

This paper describes a one-step synthesis for alloyed $Zn_xCd_{1-x}Se$ QDs at low temperature. Our results demonstrated that the composition and optical properties of these QDs can be changed by controlling the Zn^{2+}/Cd^{2+} molar ratio in the precursor solution. More importantly, in the case of the $Zn_{0.93}Cd_{0.07}Se$ QDs with white-light emission, we discovered that the intensity ratio of the band-edge to trap-state emissions was highly dependent on the reaction time and temperature. Therefore, we can balance the relative strength of the band-edge and trap-state emissions by carefully controlling the reaction time, thereby successfully producing white-light QDs. Finally, we developed a white-light, solid-state lighting device by using a 365-nm UV lamp as the pump source. We believe that this new class of alloyed QDs can potentially be applied to solid-state lighting technologies.

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Supporting Information Available: UV−vis spectra, photographs of commercial LEDs, a PL spectrum, temporal evolution, and elemental composition. This material is available free of charge via the Internet at http://pubs.acs.org.

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[36] Chan, W. C. W.; Maxwell, D. J.; Gao, X.; Bailey, R. E.; Han, M.; Nie, S. *Curr. Opin. Biotechnol.* **2002**, *13*, 40–46.