Supporting Information

Exciton quenching and ferromagnetism-to-ferrimagnetism crossover in diluted magnetic semiconducting $Zn_{1-x}Co_xO$ nanogranular nanofibers

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Figure S1. The setup of electrospinning used in our laboratory. The precision syringe pump (KDS-200, Stoelting, Wood Dale, IL) delivered the polymer precursor solution, and the voltage of electrospinning was maintained by a high-voltage power supply (Glassman High Voltage Inc. model Series FC with a positive polarity). As a high voltage was applied, the precursor solution jet accelerated towards the cathode with a 20-gauge blunt stainless steel needle tip, leading to the formation of the electrospun fiber nonwoven mats onto the AI foil placed over a grounded copper collector plate accompanied by partial solvent evaporation. Upon Co doping, the color of the buffer solution turns from colorless to pink.



Figure S2. Left, The digital photographs of the as-electrospun pure PVA/ZnO nonwoven fibers mats of (A) as-prepared, (B) sintering at T = 120 °C for one hour, and (C) 550 °C for 5 hours. The color is white, yellow, and white, respectively. Right, The digital photographs of the electrospun Co 4.4 at.% $PVA/Zn_{1-x}Co_xO$ nonwoven fibers mats of (A) as-prepared, (B) sintering at T = 120 °C for one hour, and (C) 550 °C for 5 hours. The color is white, yellow, and green, respectively. Free-standing nonwoven mats of electrospun PVA/Zn_{1-x}Co_xO fibers could also be obtained by peeling off relatively thick films from the aluminum foil. Due to the remarkable mass loss observed (for a detailed description see the text in Fig S4), the as-sintered samples undergo significant volumetric shrinkage during calcination.



Figure S3. Low- and high-magnification SEM images (A-B) of the as-electrospun PVA fibers, respectively.



Figure S4. Thermogravimetric analysis (TGA) of the as-electrospun $PVA/Zn_{1-x}Co_xO$ composite fibers with different doping level. The heating rate is at 10 K /min under static atmosphere ambient. The mass loss of the as-electrospun $PVA/Zn_{1-x}Co_xO$ composite fibers is ~ 90.8 wt % (x = 0), ~ 82.8 wt % (x = 1.8 % Co), ~ 86.0 wt % (x = 4.4% Co), and ~ 88.4 wt % (x = 7.2 % Co), respectively.



Figure S5. Typical comparison between the continuous DTA traces (well calibrated with tin) of the aselectrospun $PVA/Zn_{1-x}Co_xO$ composite fibers with Co dopant dependence (x = 0, the dashed dot curves, and x = 7.2 % Co, the solid curves) in a static atmosphere at different heating rate of (a) 5 K/min, (b) 10 K/min, (c) 15 K/min, and (d) 20 K/min, respectively.



Figure S6. PXRD patterns of the as-calcined electrospun $Zn_{1-x}Co_xO$ nanofibers with different doping level, (A) x = 0, (B) x = 1.8 % Co, (C) x = 4.4% Co, and (D) x = 7.2 % Co, respectively. Insets, lattice parameters (*a*, *c*) at different Co-doping level x.



Figure S7. PXRD data on log scale for the y-axis of the as-calcined electrospun $Zn_{1-x}Co_xO$ nanofibers with different doping level, (A) x = 0, (B) x = 1.8 % Co, (C) x = 4.4% Co, and (D) x = 7.2 % Co, respectively. There are no secondary dopant related phases patterns Insets, lattice parameters (*a*, *c*) at different Co-doping level x.



Figure S8. Evolution of the grain size (dark solid circles) and internal strain (red solid circles) of the as-calcined electrospun $Zn_{1-x}Co_xO$ nanofibers as functions of Co-doping level x.



Figure S9. (A-D) Low- and high- magnification SEM images of the as-calcined electrospun $Zn_{1-x}Co_xO$ (x = 4.4 at. %) nanofibers after mechanically fragmented.