Characterization

The crystal phase was determined by X-ray diffraction (Bruker D8 CEVANCE) using graphite monochromatized Cu-Ka ($\lambda = 1.5406$ Å) radiation. Optical properties were analyzed by using UV–vis diffuse reflectance spectra (DRS, Varian Cary 300) and photoluminescence spectra (F-7000, Hitachi, Japan) at room temperature. Fourier transformed infrared (FTIR) spectra of the samples were performed by a VERTEX-70 spectrometer, and KBr was used as a blank control. The morphology and structure of samples were studied using a scanning electron microscope (SEM, Hitachi S-4800), which was quipped for energy dispersive X-ray (EDS) analysis. Photoluminescence (PL) spectra were obtained on a Spectrofulorometer FS (Edinburgh instruments). XPS measurement was performed on a Kratos AXIS Ultra DLD spectrometer with a monochromatic AlK α X-ray source.

Computational details

Spin-polarization calculations were performed using Vienna ab initio simulation package (VASP) code based on density functional theory. The Perdew-Burke-Ernzerhof (PBE) functional was adopted to treat the electron exchange and correlation effects by the generalized gradient approximation (GGA). Three slabs with a vacuum layer of 15 Å were built: a 2 × 2 × 1 supercell of C₃N₄ monolayer and Ag doped systems. The cell parameters are a = b = 14.27 Å and c = 18.50 Å. We adopted the Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation, the DFT-D3 methodology, a cutoff energy of 450 eV, and a 3 × 3 × 1 Monkhorst-Pack grid. The optimized structures were obtained with the criterions of < 0.03 eV/Å in force and 10⁻⁵ eV in energy. C 2s²2p², N 2s²2p³, and Ag 4d¹⁰5s¹ are treated as valence electrons. We evaluated charge transfer using Bader charge analysis. The work function ($\Phi = E_{vac} - E_{fermi}$) of each system was evaluated using the difference between electrostatic potential of vacuum level (E_{vac}) and fermi energy (E_{fermi}). VASPKIT was employed to do analysis.



Fig. S1 Constant Temperature Water-bathing (EYELA PSL-1810). It adopts double compressors and strong refrigeration to achieve the required temperature faster. The temperature range is: -80 $^{\circ}$ C - 0 $^{\circ}$ C, and we use anhydrous ethanol as refrigerants. Therefore, we can easily control the temperature at -60 $^{\circ}$ C with stirring for 4 h.



Fig. S2. XRD patterns of g-CN and a series of Ag/CN composites.



Fig. S3. Finger spectrum of Ag/CN(-60)



Fig. S4. Mapping picture of Ag/CN(-60)



Fig. S5. EDX spectrum of Ag/CN(-60)



Fig. S6. Plots of $(Ahv)^{1/2}$ vs. hv spectra



Fig. S7. Mott-Schottky plots of three samples



Fig. S8. The estimated band gap distribution.