Update to Electronic Supplementary Information on 09/08/22:

For the sake of concentrating on the main scientific findings instead of the specific data, line style was used when the authors created some of the figures in the original paper. However, for Fig. 3 and Fig. 11D, this plotting style might make the curves corresponding to different samples seem to be identical to each other, which is not the case. On the other hand, the data shift that changed the plot scale in Fig. 3 might also contribute to this confusion. Therefore, here we re-express these data in a more detailed manner, i.e. plotting directly the raw data points rather than the curves in the same plot scale (no data shift) to clearly address the difference between the data set of different samples.

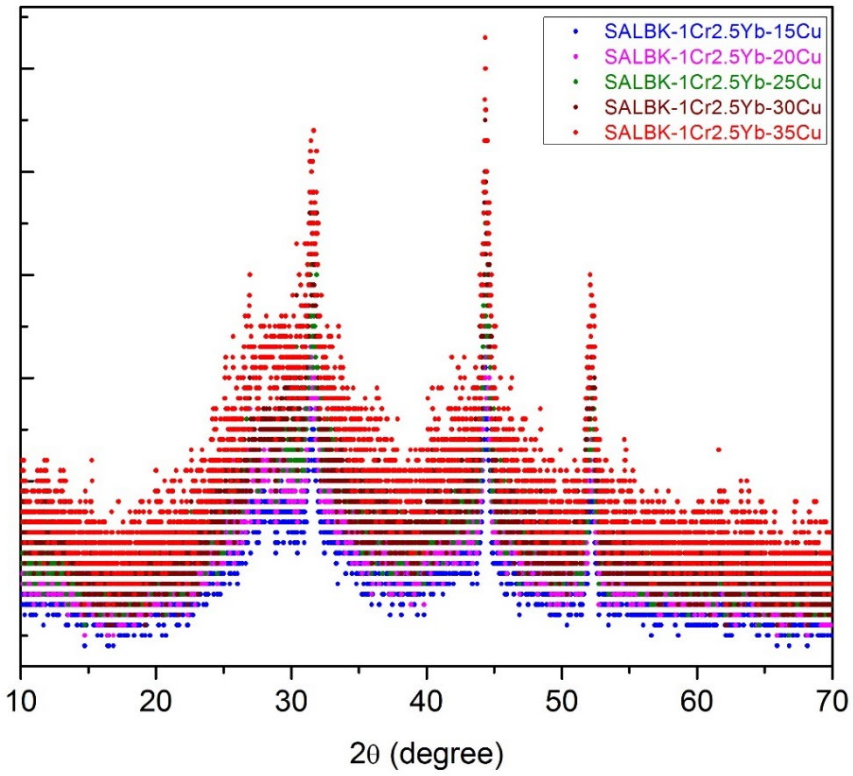

Figure S1: The more detailed plot of all the XRD data in Fig. 3 (except the black one) in the same plotting frame, plotted in points within the same plot scale.

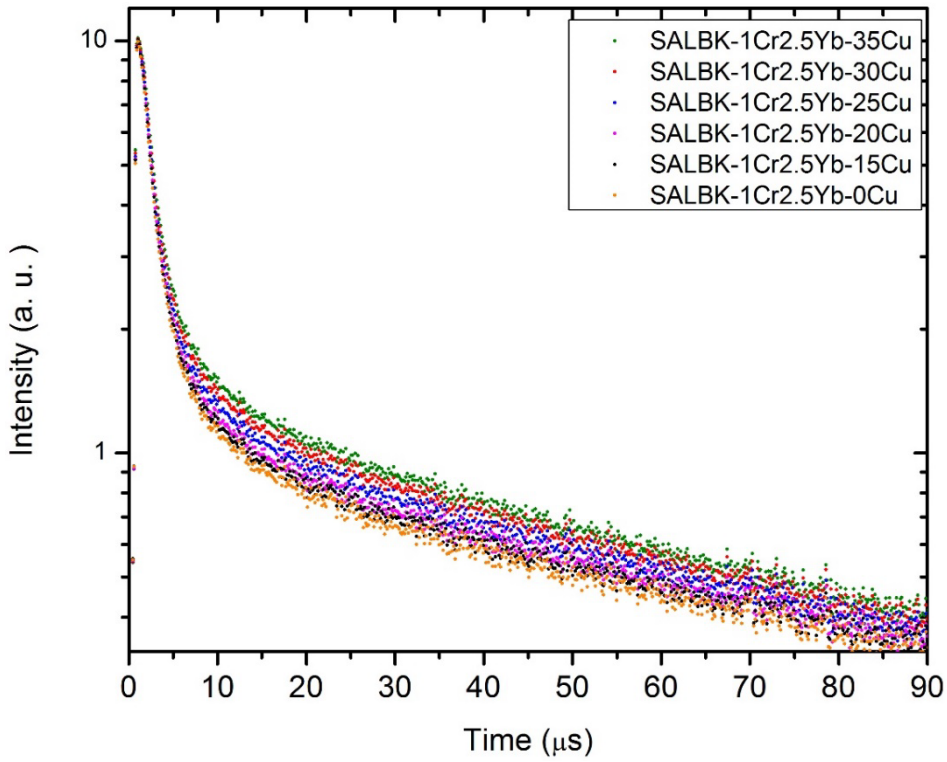

Figure S2: The more detailed plot of all decay lifetime data in Fig. 11D, plotted in points.

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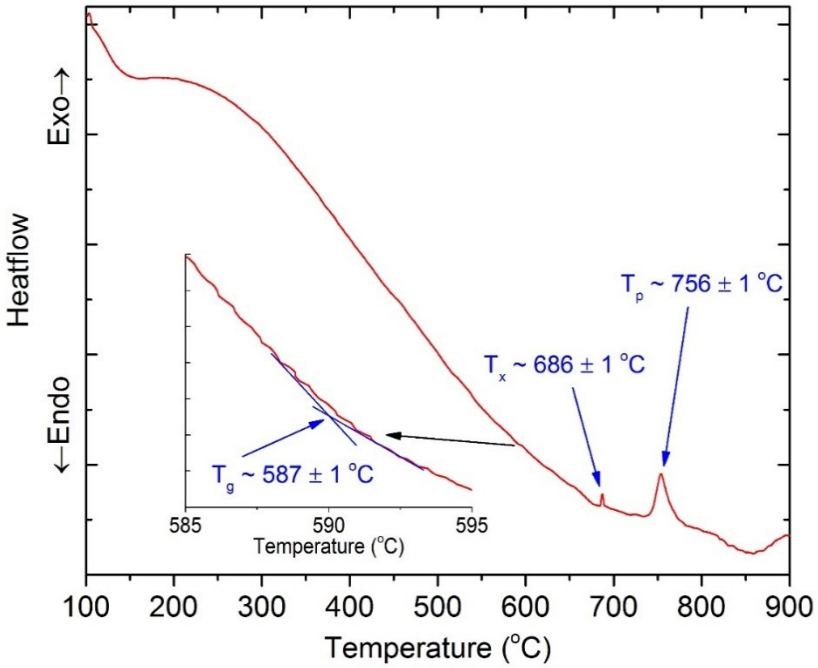

Fig. S1 DTA analysis of $45SiO_2$–$20Al_2O_3$–$12.5LaF_3$–$10BaF_2$–$9K_2O$–$1Cr_2O_3$–$2.5Yb_2O_3$ (SALBK) glass sample.

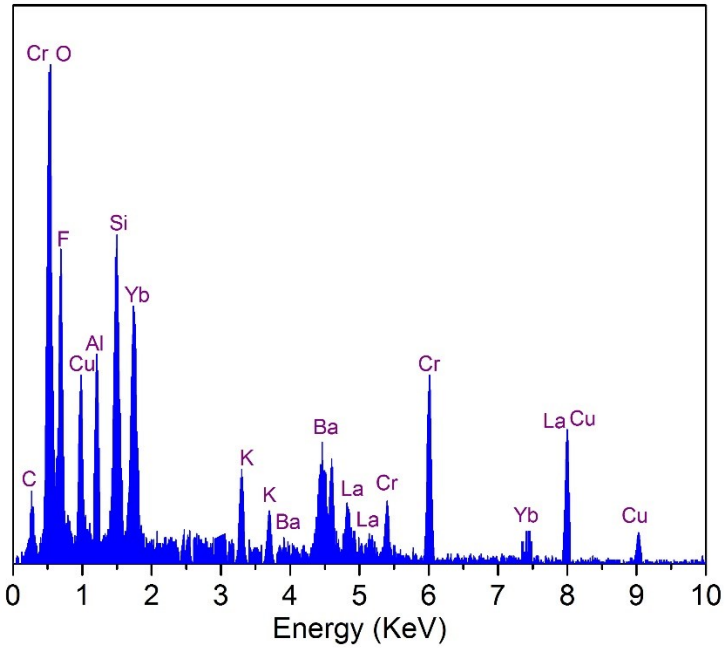

Fig. S2 EDS analysis of SALBK-1Cr2.5Yb-35Cu glass sample.

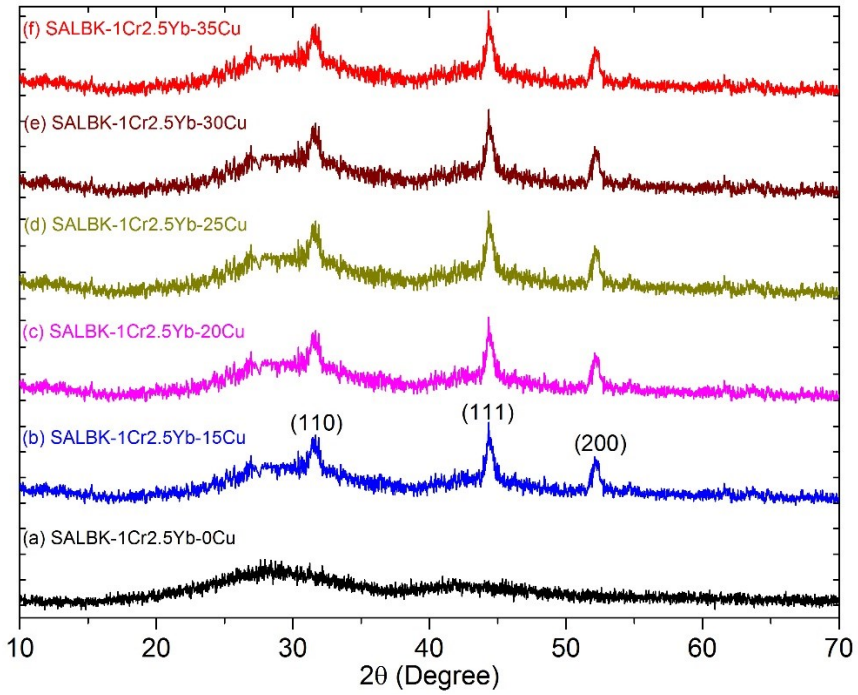

Fig. S3 XRD analysis of SALBK-1Cr2.5Yb-0Cu, SALBK-1Cr2.5Yb-15Cu, SALBK-1Cr2.5Yb-20Cu, SALBK-1Cr2.5Yb-25Cu, SALBK-1Cr2.5Yb-30Cu, and SALBK-1Cr2.5Yb-35Cu glass samples.

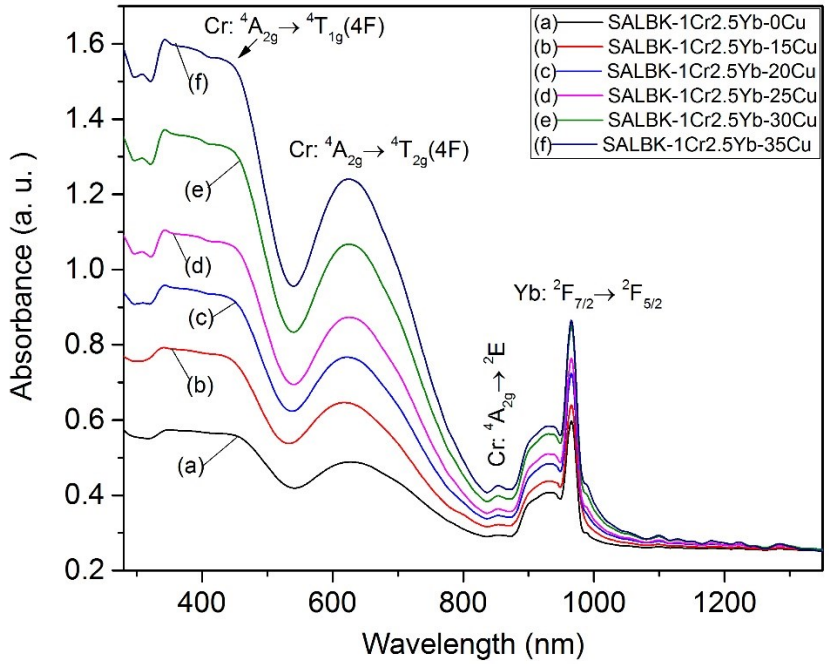

Fig. S4 Absorption spectra of SALBK-1Cr2.5Yb-0Cu, SALBK-1Cr2.5Yb-15Cu, SALBK-1Cr2.5Yb-20Cu, SALBK-1Cr2.5Yb-25Cu, SALBK-1Cr2.5Yb-30Cu, and SALBK-1Cr2.5Yb-35Cu glass samples.

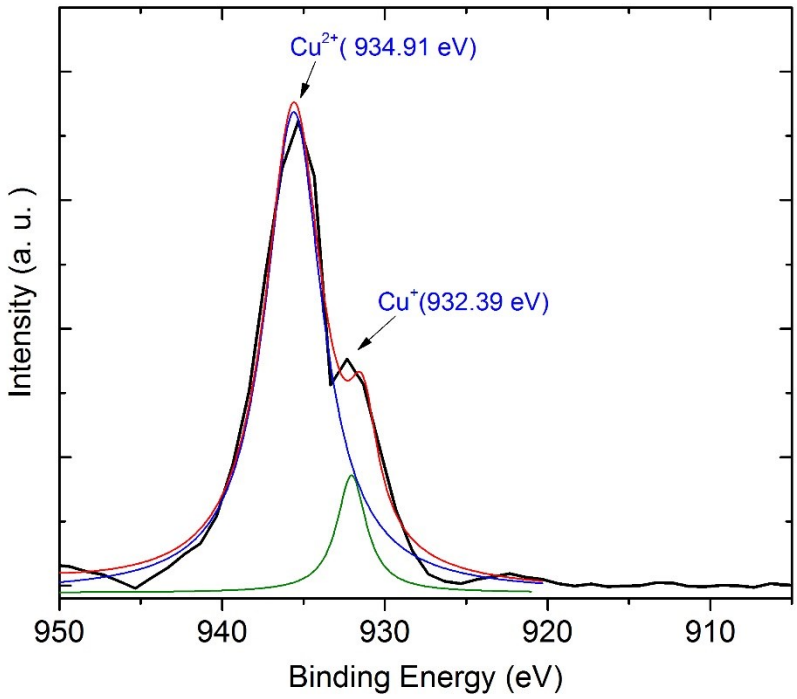

Fig. S5 XPS spectra of SABLK-15Cu glass sample.

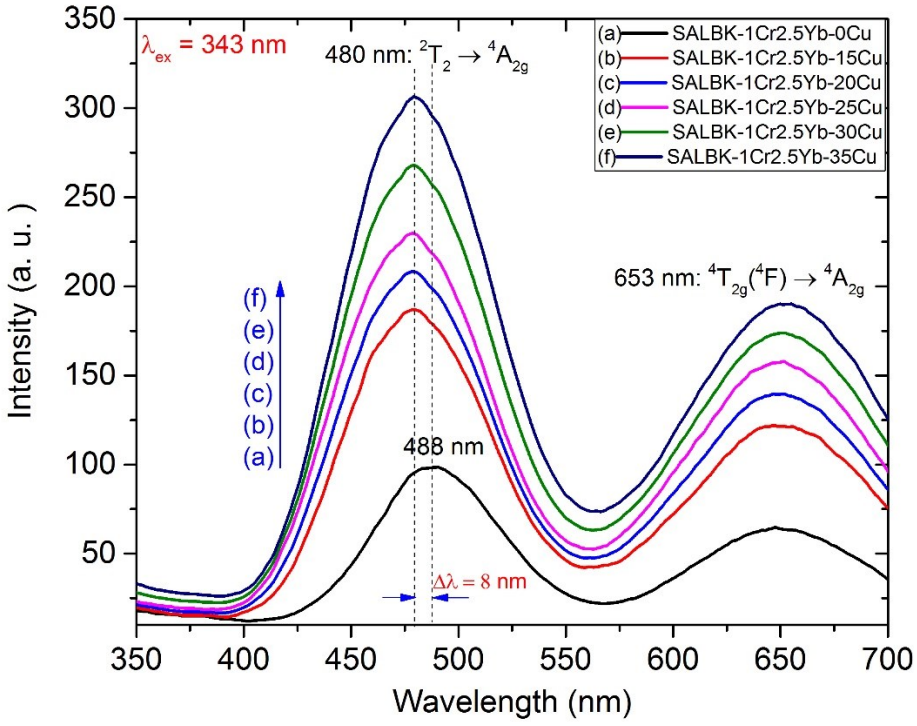

Fig. S6 Visible emission spectra of SALBK-1Cr2.5Yb-0Cu, SALBK-1Cr2.5Yb-15Cu, SALBK-1Cr2.5Yb-20Cu, SALBK-1Cr2.5Yb-25Cu, SALBK-1Cr2.5Yb-30Cu, and SALBK-1Cr2.5Yb-35Cu glass samples under excitation 343 nm.

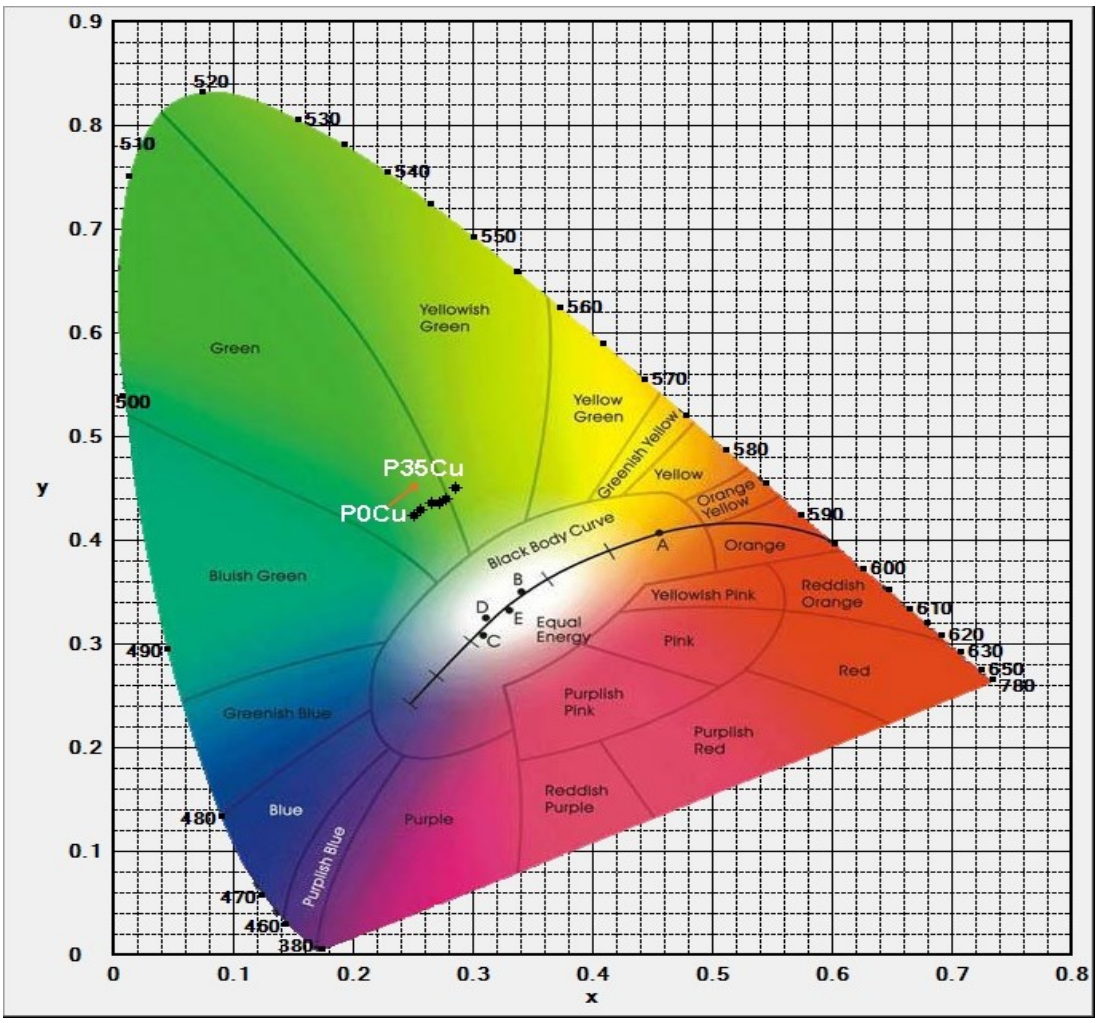

Fig. S7 CIE 1931 (x; y) chromaticity coordinates for luminescence of Cr^{3+}/Yb^{3+} co-doped in SALBK-1Cr2.5Yb-0Cu, SALBK-1Cr2.5Yb-15Cu, SALBK-1Cr2.5Yb-20Cu, SALBK-1Cr2.5Yb-25Cu, SALBK-1Cr2.5Yb-30Cu, and SALBK-1Cr2.5Yb-35Cu glass samples.

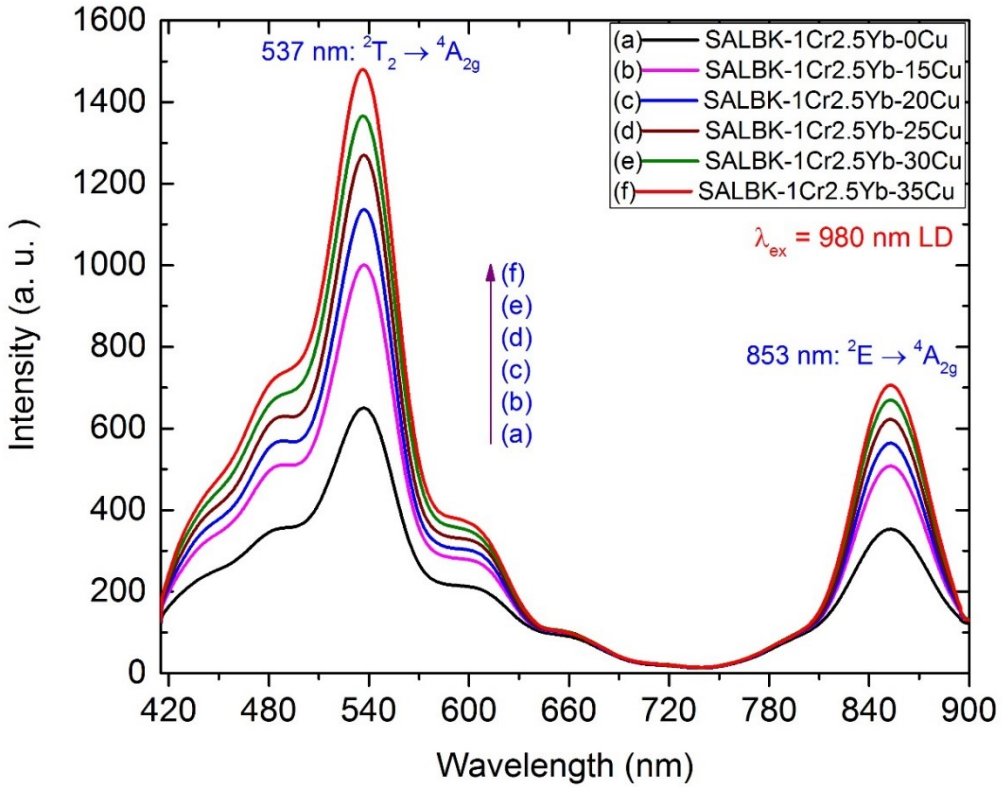

Fig. S8 UC spectra of Cr^{3+}/Yb^{3+} co-doped in SALBK-1Cr2.5Yb-0Cu, SALBK-1Cr2.5Yb-15Cu, SALBK-1Cr2.5Yb-20Cu, SALBK-1Cr2.5Yb-25Cu, SALBK-1Cr2.5Yb-30Cu, and SALBK-1Cr2.5Yb-35Cu glass samples under excitation 980 nm LD.

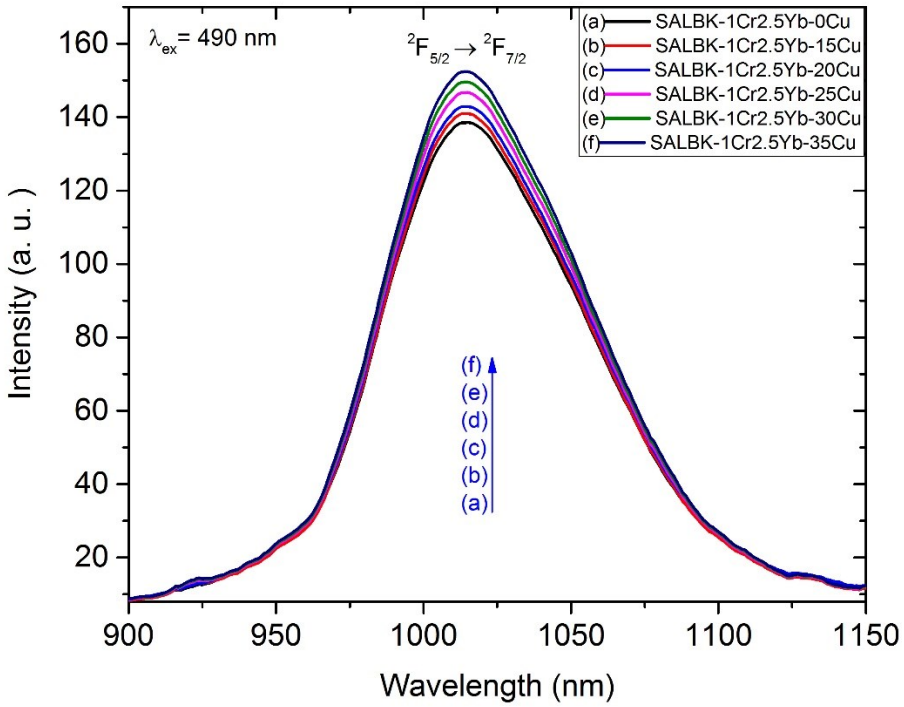

Fig. S9 NIR emission spectra of Cr^{3+}/Yb^{3+} co-doped in SALBK-1Cr2.5Yb-0Cu, SALBK-1Cr2.5Yb-15Cu, SALBK-1Cr2.5Yb-20Cu, SALBK-1Cr2.5Yb-25Cu, SALBK-1Cr2.5Yb-30Cu, and SALBK-1Cr2.5Yb-35Cu glass samples, excited by 490 nm.

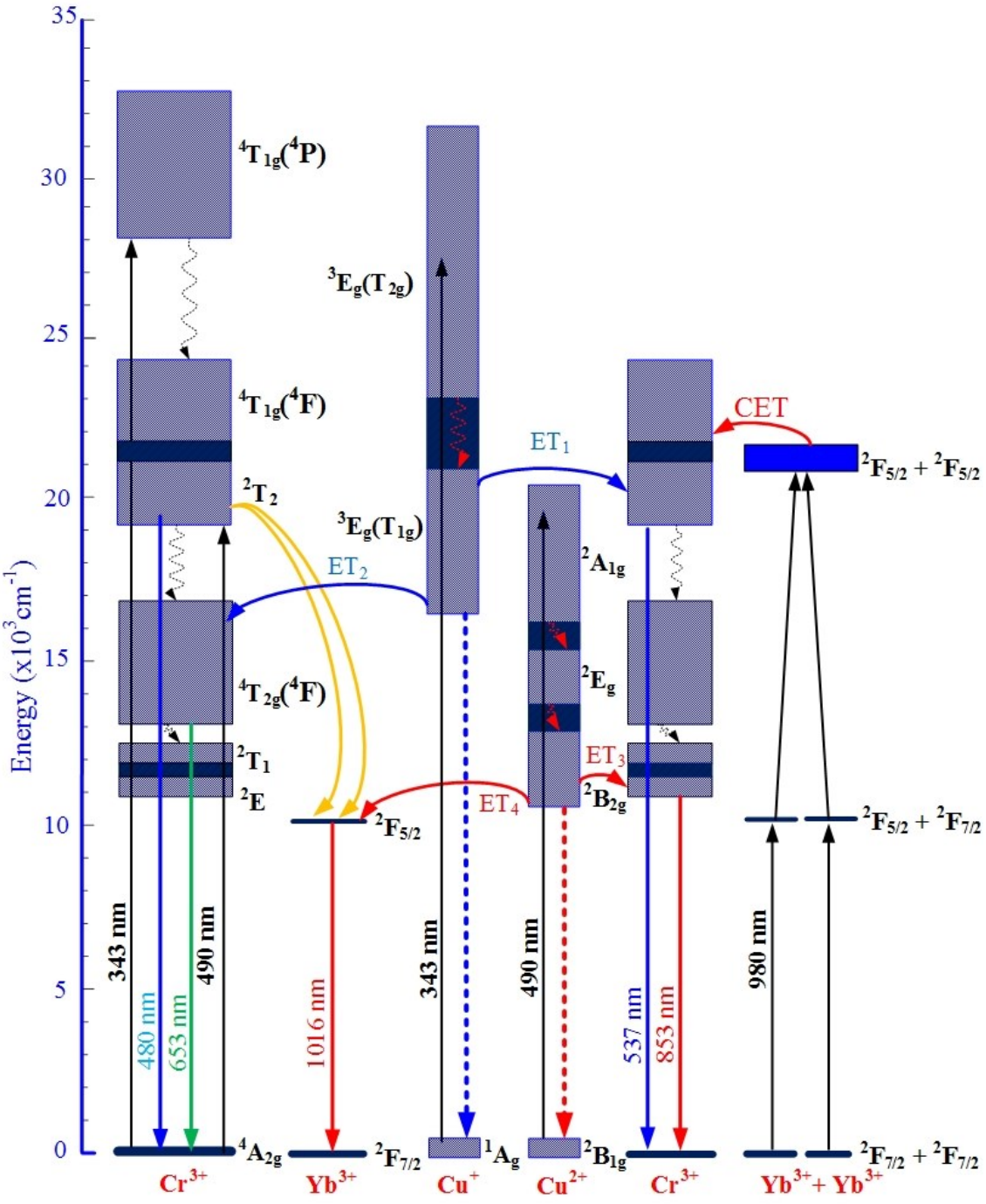

Fig. S10 Mechanism ET processes for the visible, UC, and NIR luminescence of Cr^{3+}/Yb^{3+} co-doped under excitations 343, 490, and 980 nm LD.

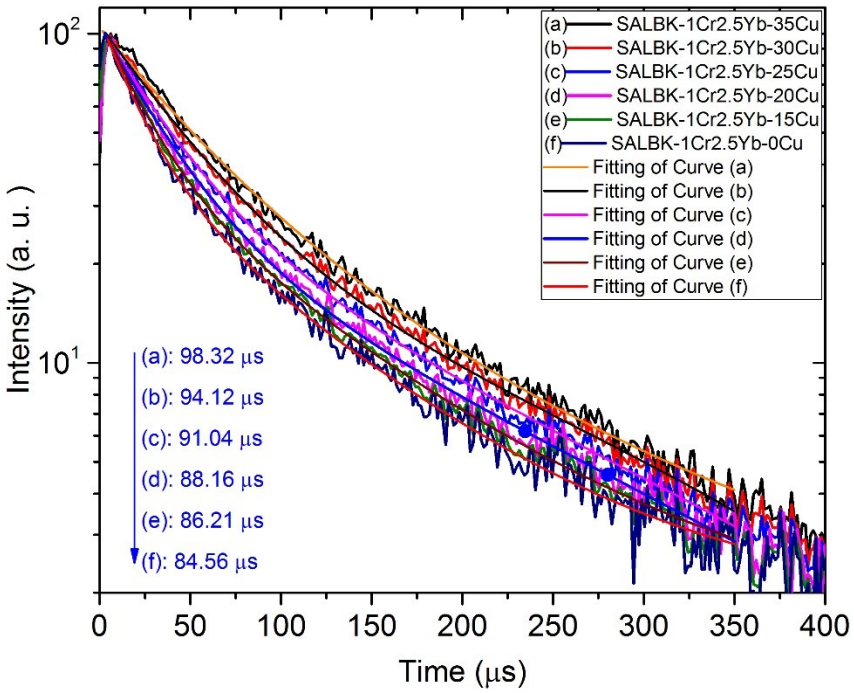

Fig. S11a Decay lifetimes curves of Cr^{3+} at 537 nm in SALBK-0Cu, SALBK-15Cu, SALBK-20Cu, SALBK-25Cu, SALBK-30Cu, and SALBK-35Cu glass samples, under excitation 980 nm.

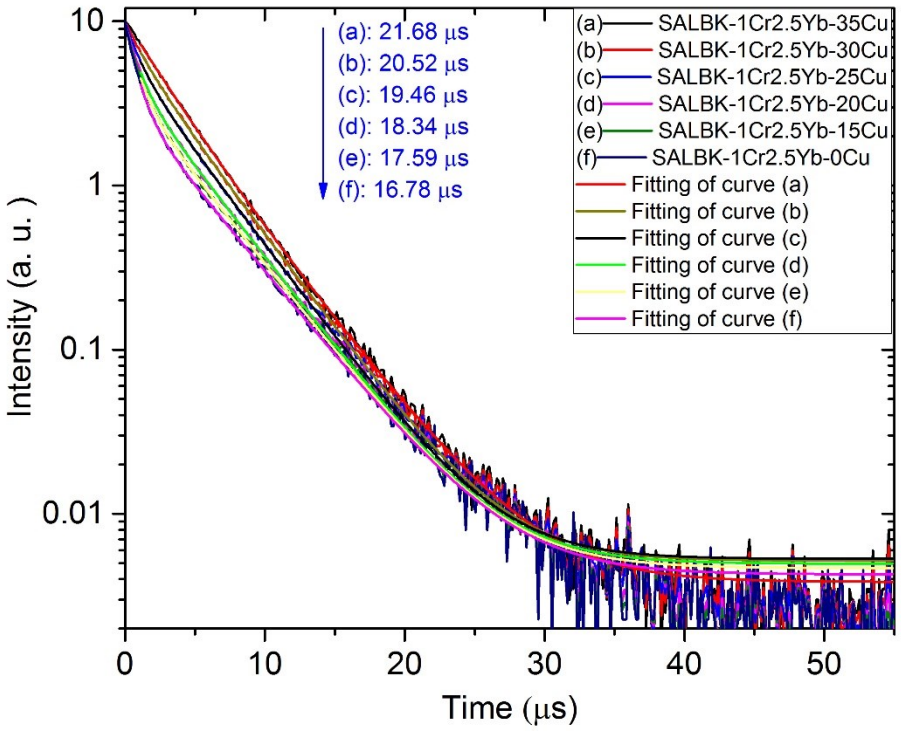

Fig. S11b Decay lifetimes curves of Cr^{3+} at 653 nm in SALBK-0Cu, SALBK-15Cu, SALBK-20Cu, SALBK-25Cu, SALBK-30Cu, and SALBK-35Cu glass samples, under excitation 343 nm.

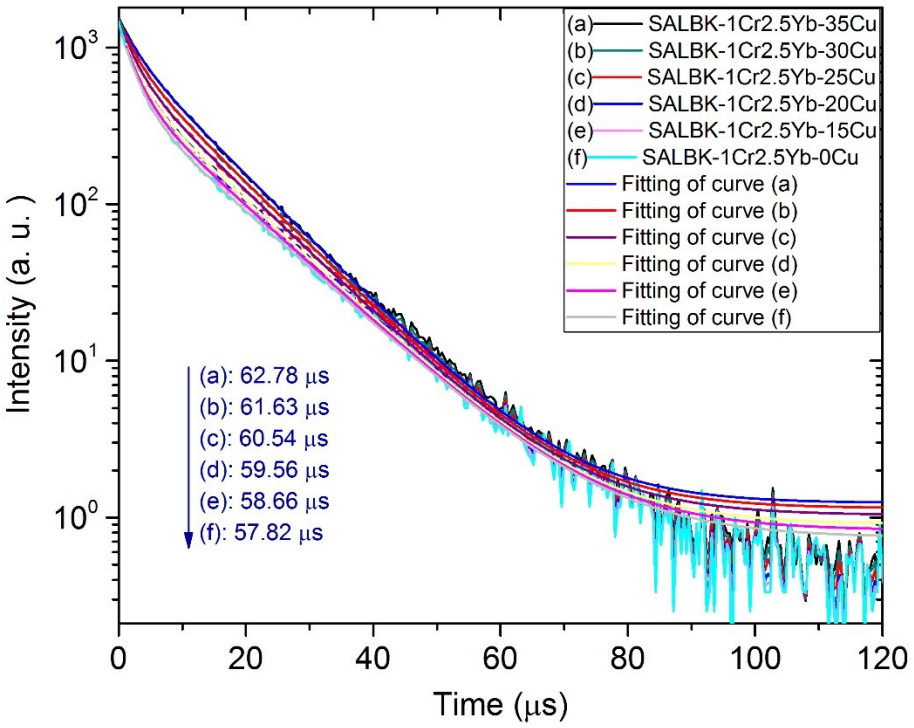

Fig. S11c Decay lifetimes curves of Cr^{3+} at 853 nm in SALBK-0Cu, SALBK-15Cu, SALBK-20Cu, SALBK-25Cu, SALBK-30Cu, and SALBK-35Cu glass samples, under excitation 980 nm LD.

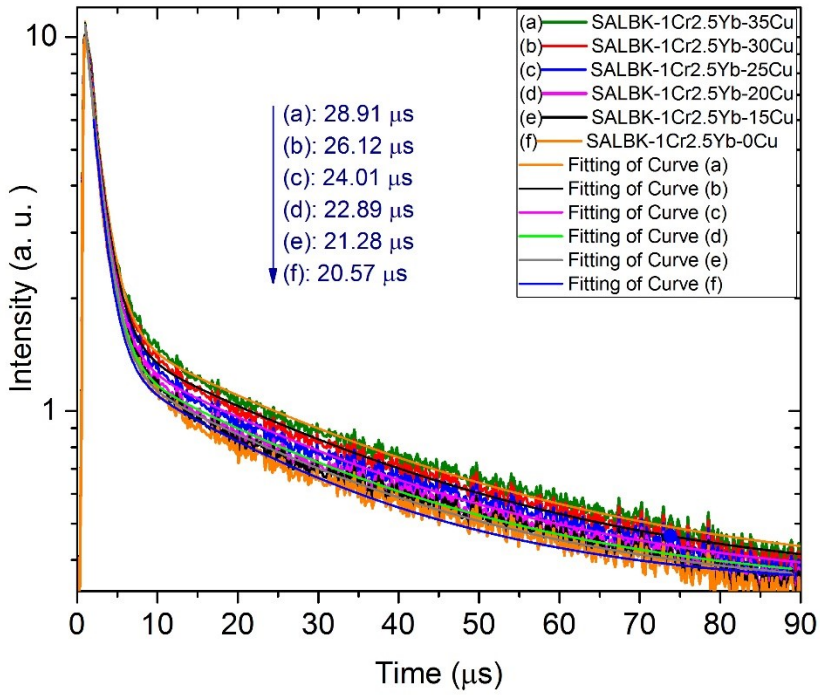

Fig. S11d Decay lifetimes curves of Yb^{3+} at 1016 nm in SALBK-0Cu, SALBK-15Cu, SALBK-20Cu, SALBK-25Cu, SALBK-30Cu, and SALBK-35Cu glass samples, under excitation 490 nm.

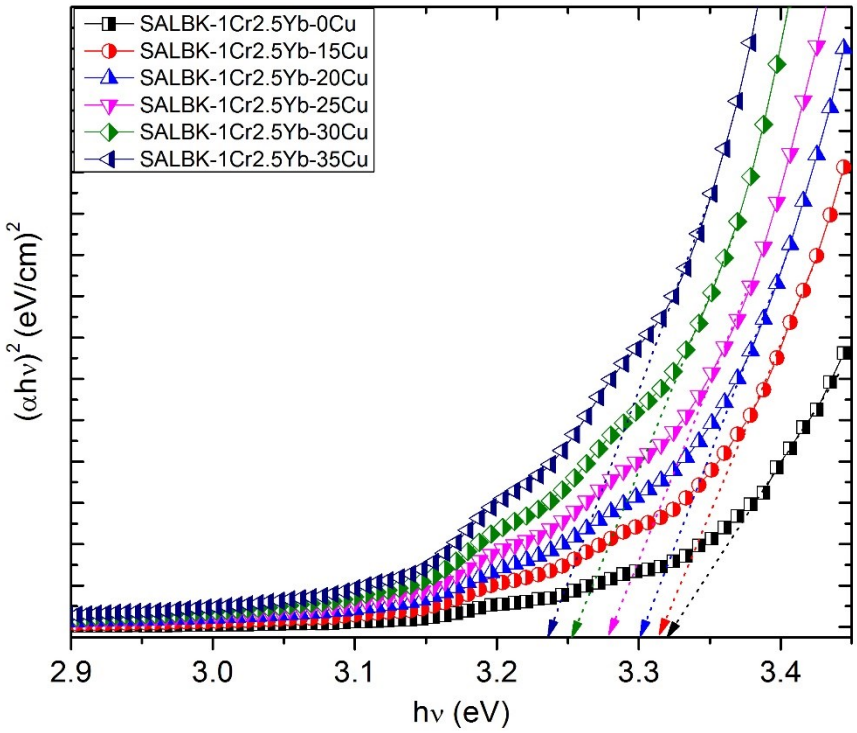

Fig. S12 Plot of (hν) versus $(\alpha h\nu)^2$ for estimating the E_g of SALBK-1Cr2.5Yb-0Cu, SALBK-1Cr2.5Yb-15Cu, SALBK-1Cr2.5Yb-20Cu, SALBK-1Cr2.5Yb-25Cu, SALBK-1Cr2.5Yb-30Cu, and SALBK-1Cr2.5Yb-35Cu glass samples.

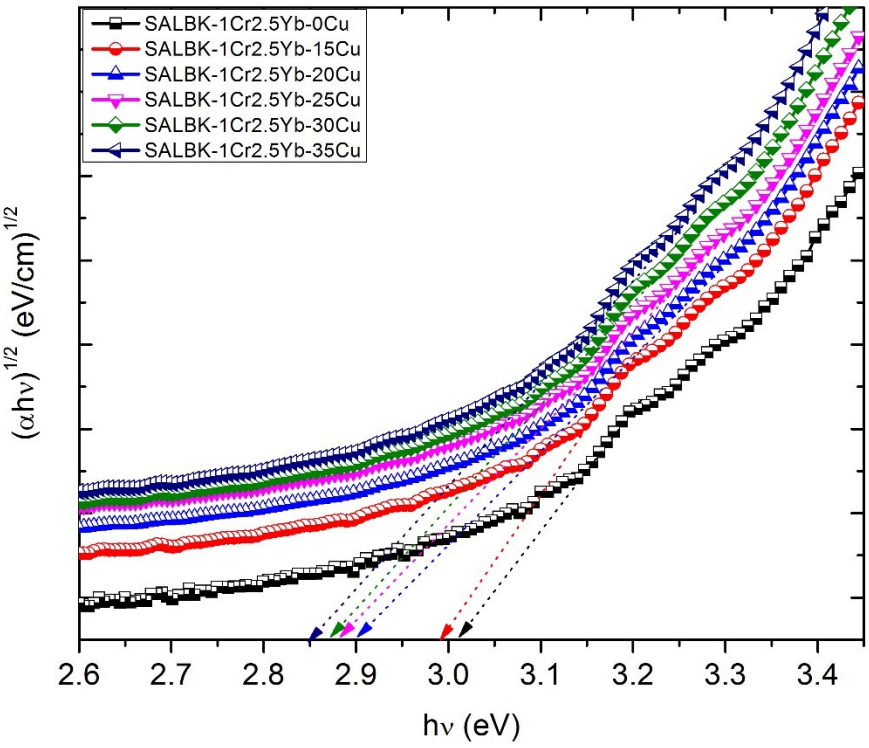

Fig. S13 Plot of (hν) versus $(\alpha h\nu)^{1/2}$ for estimating the E_g of SALBK-1Cr2.5Yb-0Cu, SALBK-1Cr2.5Yb-15Cu, SALBK-1Cr2.5Yb-20Cu, SALBK-1Cr2.5Yb-25Cu, SALBK-1Cr2.5Yb-30Cu, and SALBK-1Cr2.5Yb-35Cu glass samples.