

Supplementary Information for
**RIXS as a probe of $J_{\text{eff}} = 1/2$ state in $3d$
transition-metal oxide CuAl_2O_4**

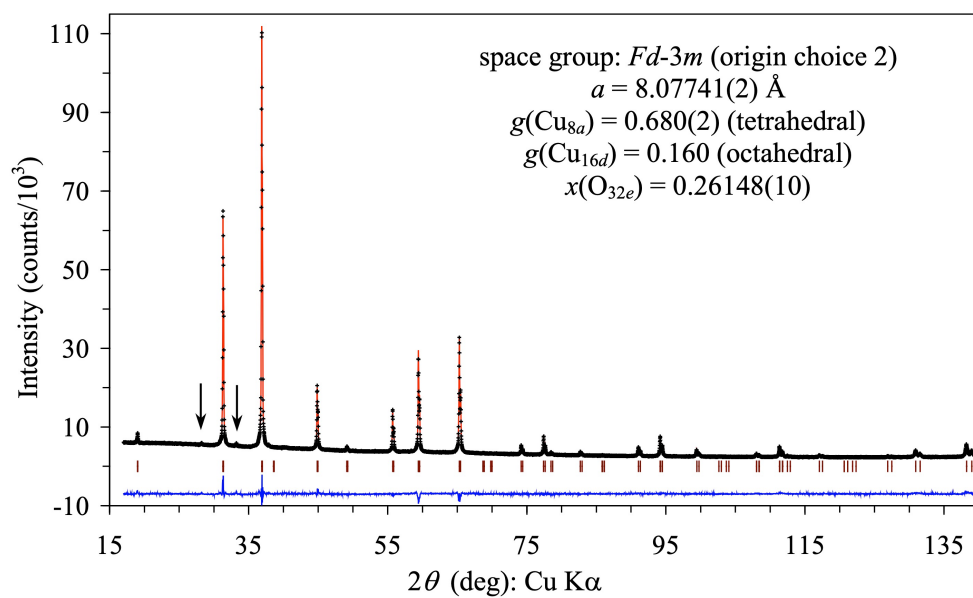
H. Y. Huang, A. Singh, C. I. Wu, J. D. Xie, J. Okamoto, A. A. Belik
E. Kurmaev, A. Fujimori, C. T. Chen, S. V. Streltsov[†], D. J. Huang^{*}

Correspondence to:

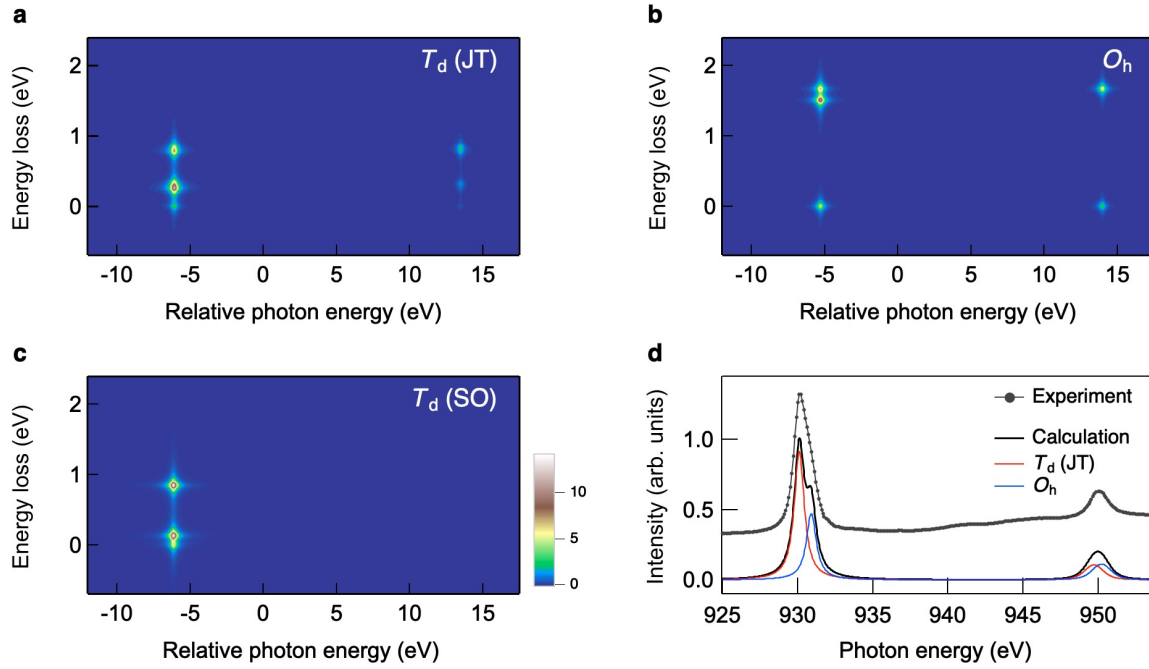
[†] streltsov@imp.uran.ru

^{*} djhuang@nsrrc.org.tw

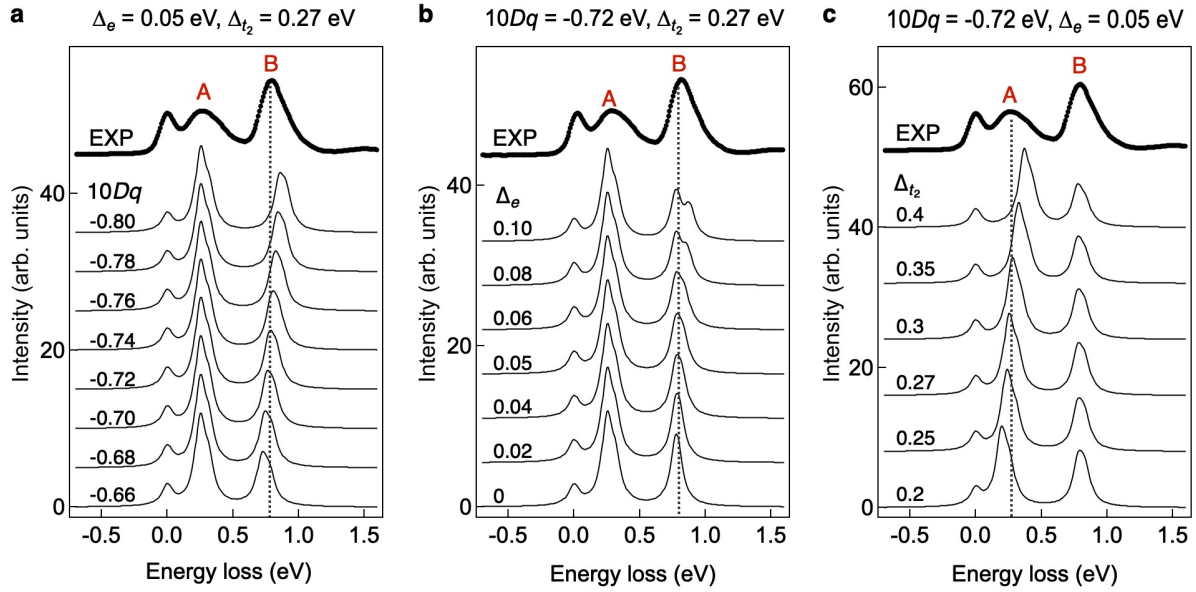
January 17, 2022



Supplementary Figure 1: The experimental (black crosses), calculated (red line), and difference (blue line at the bottom) X-ray diffraction patterns of CuAl_2O_4 ($Z = 8$). The tick marks show possible Bragg reflection positions. Arrows show small contributions from the Cu $K\beta_1$ radiation. The main refinement results are given on the figure; g is the occupation factor.



Supplementary Figure 2: Multiplet calculations of Cu^{2+} . (a), (b) & (c) Cu L -edge RIXS intensity maps of Jahn-Teller distorted tetrahedron (T_d (JT)), octahedron (O_h), and spin-orbital entangle tetrahedron (T_d (SO)) calculated by QUANTY. Because the experiments were done with a powder sample, here each RIXS calculation is an average of the results obtained from three possible geometries (scattering planes on XY, YZ, and ZX). (d) The calculated XAS spectra compared to the experimental data taken with total electron yield (grey filled circle). The resonant energies of XAS calculations are shifted according to the experimental RIXS results as shown in Fig. 2. The black line is the summation of 62.5 % T_d (JT) and 37.5 % O_h .



Supplementary Figure 3: Multiplet calculations of Jahn-Teller distorted tetrahedron Cu^{2+} compared with experimental RIXS spectrum. (a) Calculations with fixed $\Delta_e = 50$ meV and $\Delta_{t_2} = 270$ meV with different $10Dq$ values in eV. (b) Calculations with fixed $10Dq = -0.72$ eV and $\Delta_{t_2} = 270$ meV with several distortion Δ_e values in eV. (c) Calculations with fixed $10Dq = -0.72$ eV and $\Delta_e = 50$ meV with several distortion Δ_{t_2} values in eV. The experimental RIXS spectrum (EXP) is measured at incident photon energy 930 eV. The dashed lines give a guide to the eye in comparison with the experimental data.