## SUPPLEMENTARY INFORMATION TO

## EPR, ENDOR AND HYSCORE STUDY OF X-RAY INDUCED CENTRES IN K<sub>2</sub>YF<sub>5</sub> THERMOLUMINESCENT PHOSPHORS

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Cross peak linking the triple-quantum (tq) frequencies in the <sup>39</sup>K HYSCORE experiment



**Fig. S1** [-8 - -5, 1.5 – 3.5] MHz region from the experimental HYSCORE spectrum of K<sub>2</sub>YF<sub>5</sub>:Ce (0.1%). Magnetic field  $B_0$ =343.95 mT along the *a*-axis corresponds to the C1 centre EPR line position. The cross peak between the tq frequencies (- $v_{58}$ ,  $v_{14}$ ) of the type-I <sup>39</sup>K interaction is shown.

## <sup>19</sup>F HYSCORE

As mentioned in the main text, the weak 19F modulations are largely surppressed by the strong  $^{39}$ K modulations in the HYSCORE spectrum. As a consequence, only weak 19F cross peaks are observed in the area where we expect contributions for interactions 2, 3.1 and 3.2. The  $^{19}$ F signals due to interaction 1 (hyperfine splitting in this observer position ~12 MHz) are not observed.



Fig S2. <sup>19</sup>F HYSCORE spectrum of  $K_2$ YF<sub>5</sub>:Ce (0.1%). Magnetic field B=343.95 mT along the *a*-axis corresponds to the C1 centre EPR line position.