SUPPLEMENTARY INFORMATION TO

EPR, ENDOR AND HYSCORE STUDY OF X-RAY INDUCED CENTRES IN K$_2$YF$_5$ THERMOLUMINESCENT PHOSPHORS

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Cross peak linking the triple-quantum (tq) frequencies in the $^{39}$K HYSCORE experiment

![HYSCORE Spectrum](image)

**Fig. S1** [-8 - -5, 1.5 – 3.5] MHz region from the experimental HYSCORE spectrum of K$_2$YF$_5$: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 $^{39}$K interaction is shown.

$^{19}$F HYSCORE

As mentioned in the main text, the weak 19F modulations are largely suppressed 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.

![HYSCORE Spectrum](image)

**Fig S2.** $^{19}$F HYSCORE spectrum of K$_2$YF$_5$:Ce (0.1%). Magnetic field $B$=343.95 mT along the $a$-axis corresponds to the C1 centre EPR line position.