

# Supplementary Information for 'Dynamic Jahn-Teller effect in the strong spin-orbit coupling regime'

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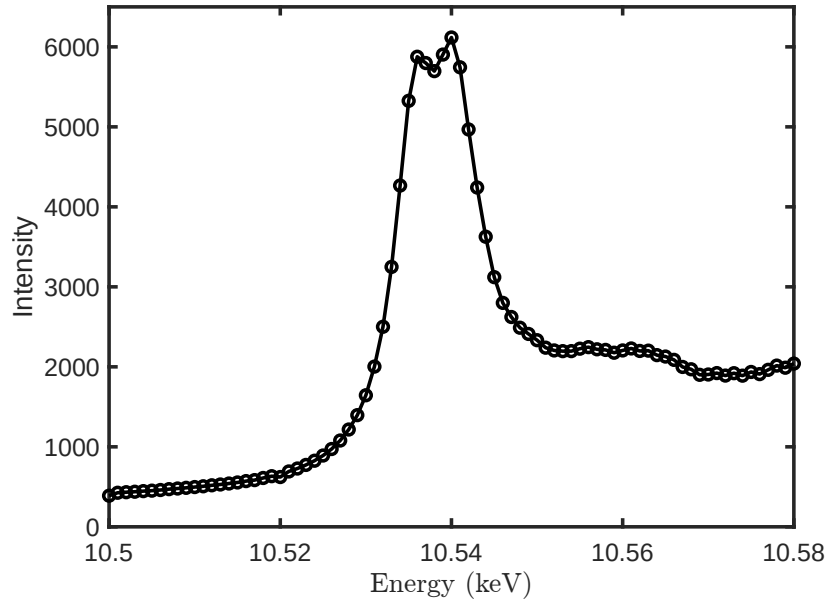
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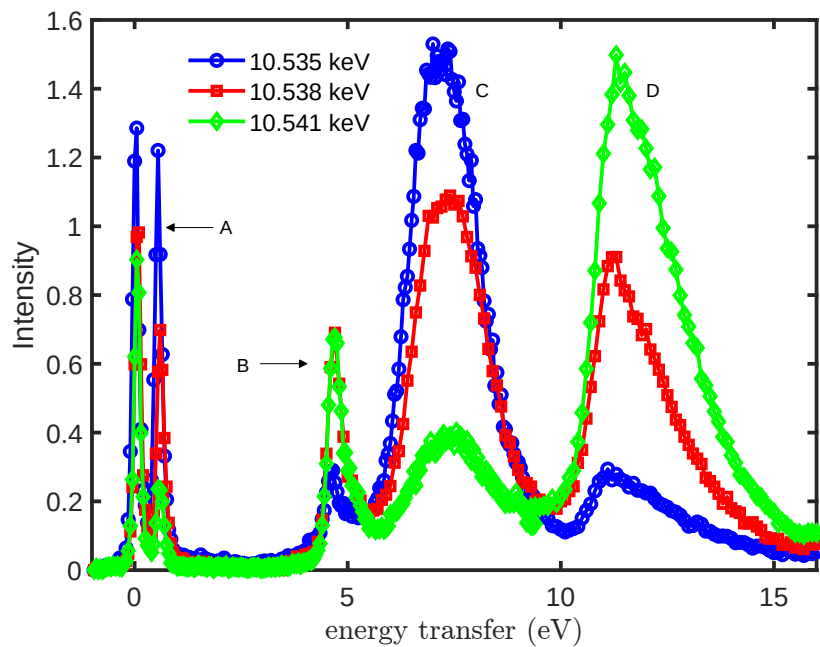
## Supplementary Note 1. RHENIUM $L_3$ EDGE RIXS



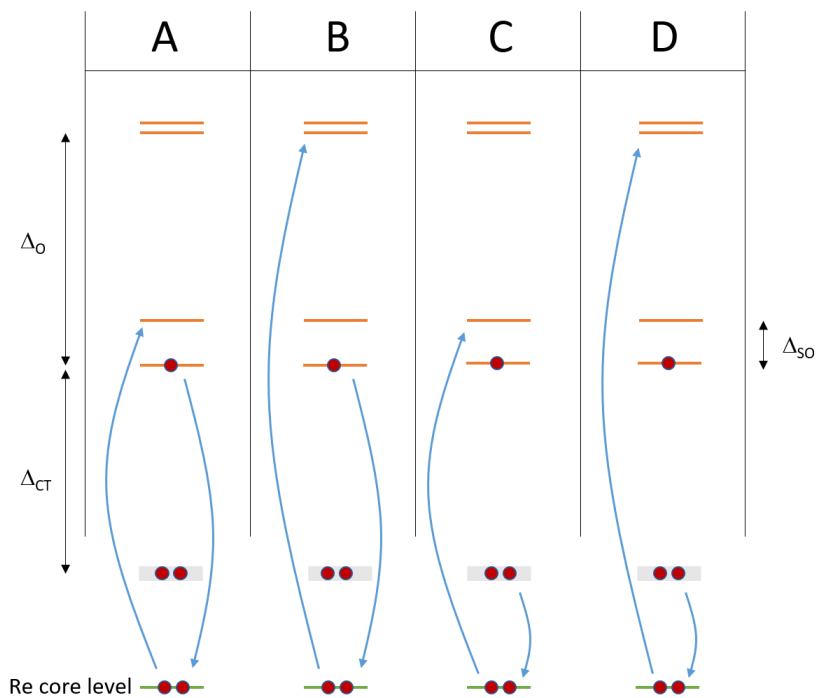
Supplementary Figure 1. Fluorescence scan on the  $L_3$  edge of Re ion.

An energy scan across the Re  $L_3$  resonance is shown in Supplementary Figure 1 and exhibits two maxima, at 10.535 keV and 10.541 keV. We have performed RIXS measurements at those two energies and an additional one at 10.538 keV (Supplementary Figure 6).

All three incident energies show qualitatively the same features in their line-shape. Alongside the elastic line, there are four features that can be associated with a specific resonant process (see Supplementary Figure 3). Features *A* and *B* can be assigned to crystal-field levels separated from the ground state by  $\Delta_{SO}$  and  $\Delta_0$ . Features *C* and *D* reflect similar crystal-field level scheme, with the involvement of the charge-transfer gap  $\Delta_{CT}$ .

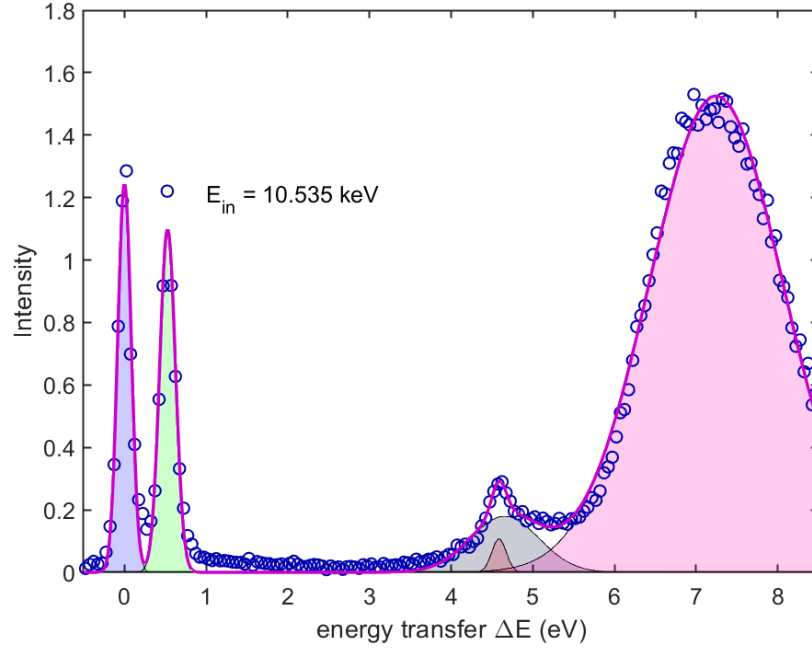


Supplementary Figure 2. Energy loss scan at three incident energies.

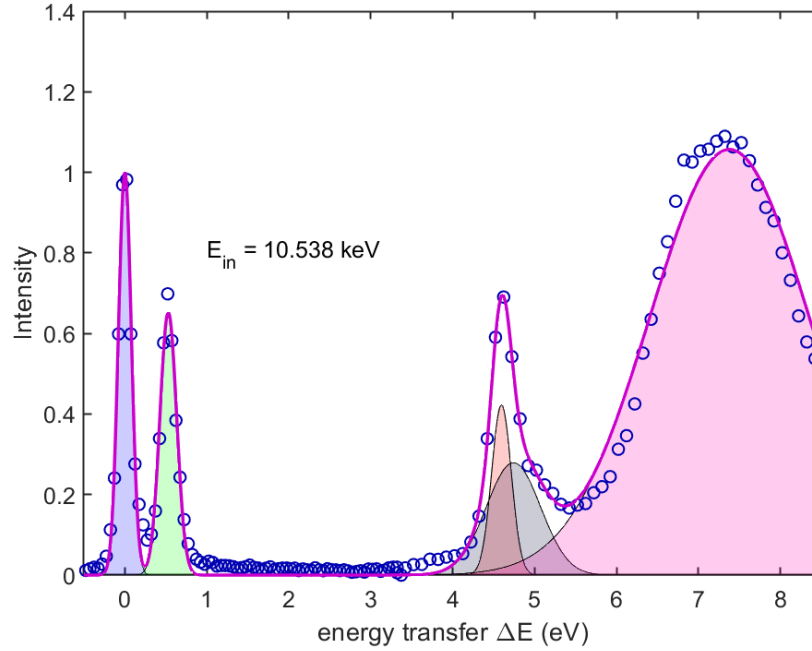


Supplementary Figure 3. A scheme of Re localized levels, including the predominantly oxygen *p*-orbital states separated by a charge-transfer gap  $\Delta_{CT}$  from the lowest occupied *d*-orbital. Each process is associated with a feature observed in Re RIXS spectra (Figure S2).

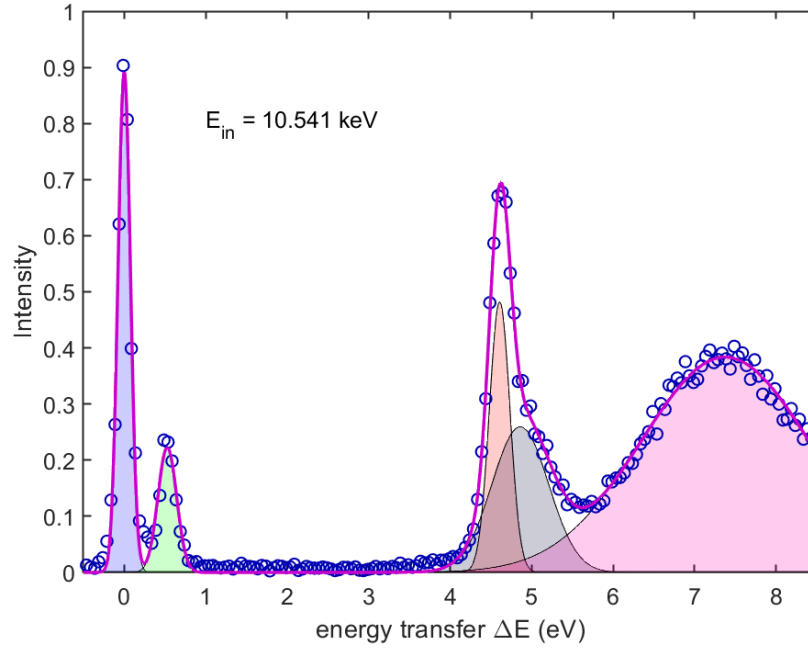
For all three incident energies we can describe the relevant energy range for crystal-field determination using 5 Gaussian modes. For lower incident energies the feature around 7.4 eV is dominating the line-shape, and its irregular shape significantly affects the quality of the fit. Nevertheless, as can be seen from Supplementary Table 1, its effect is limited to  $e_g$  levels, since the value of  $\Delta_{SO}$  extracted from the fit is the same in all three cases. For  $E_{in} = 10.541$  keV the relative intensities between  $B$  and  $C$  features are the most favorable for the determination of the positions of  $e_g$  levels.



Supplementary Figure 4. Re RIXS line-shape described with 5 Gaussian modes for  $E = 10.535$  keV.



Supplementary Figure 5. Re RIXS line-shape described with 5 Gaussian modes for  $E = 10.538$  keV.



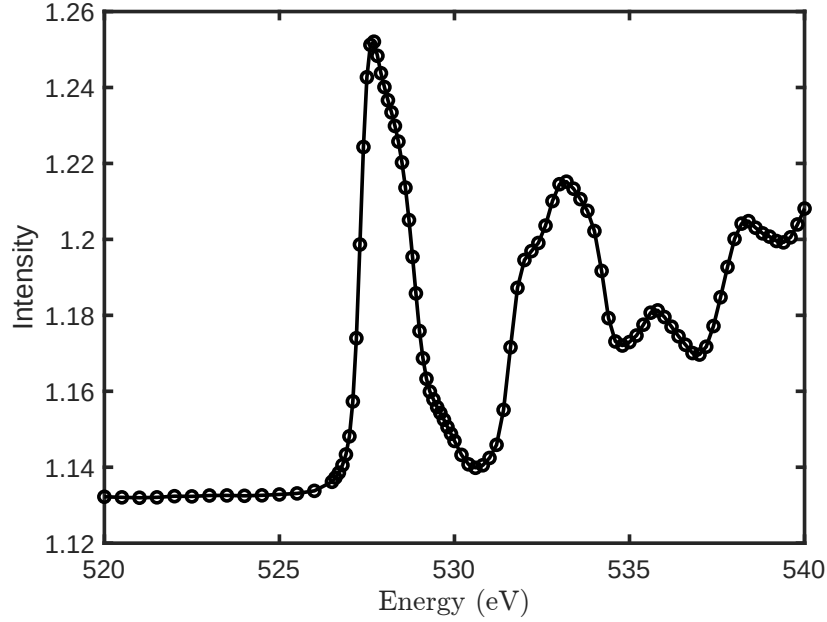
Supplementary Figure 6. Re RIXS line-shape described with 5 Gaussian modes for  $E = 10.541$  keV.

| E (meV)               | elastic | SO  | $E_{SO} - E_{elas}$ | $e_g^1$ | $e_g^2$ | CT   |
|-----------------------|---------|-----|---------------------|---------|---------|------|
| $E_{in} = 10.535$ keV | 26      | 554 | 528                 | 4613    | 4684    | 7271 |
| $E_{in} = 10.538$ keV | 77      | 609 | 532                 | 4673    | 4819    | 7456 |
| $E_{in} = 10.541$ keV | 63      | 591 | 528                 | 4671    | 4924    | 7438 |

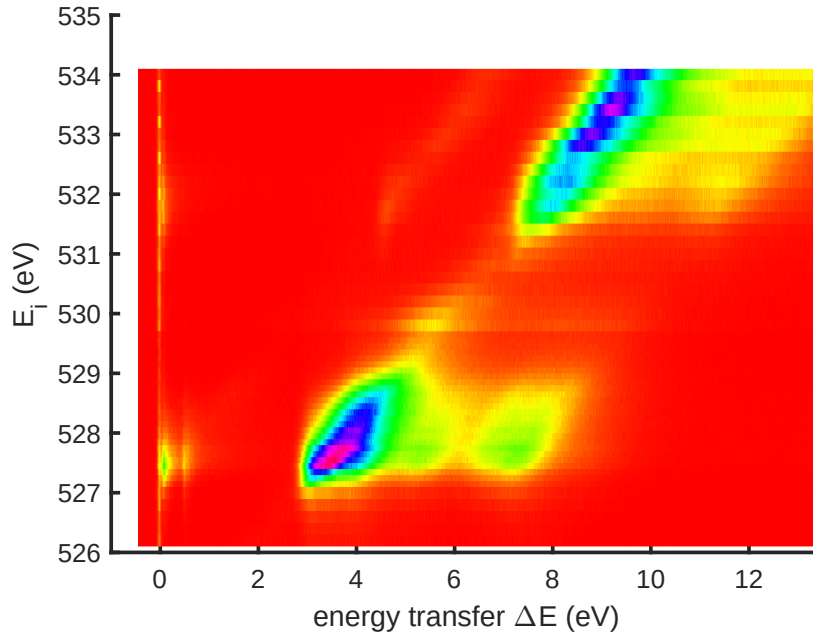
Supplementary Table 1. Energies of 5 Gaussian modes for three different incident energies.

## Supplementary Note 2. OXYGEN *K* EDGE RIXS

An energy scan across the O *K* resonance is shown in Supplementary Figure 7 and exhibits a maximum at 527.6 eV. A full map of energy transfer with respect to incident energy shown in Supplementary Figure 8 indicates narrow, incident-energy-independent features below 1 eV which can be associated with localized *d-d* excitations while above 2.5 eV a strong fluorescence signal appears.



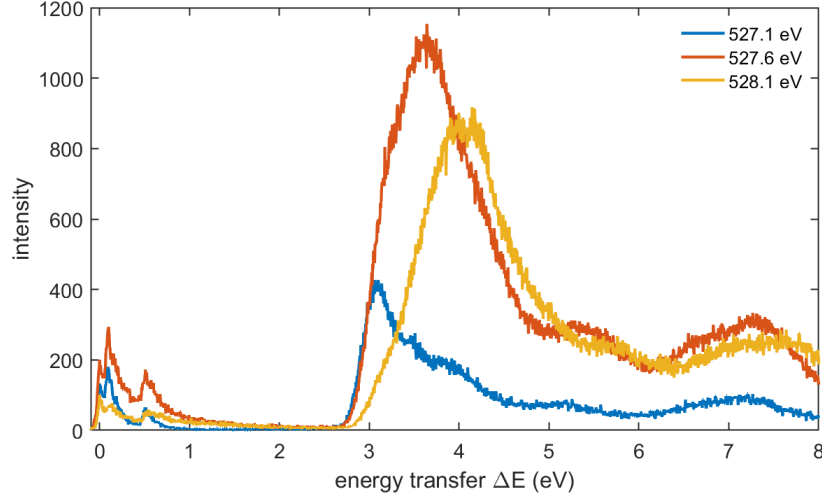
Supplementary Figure 7. Total fluorescence yield x-ray absorption scan on the oxygen *K* edge.



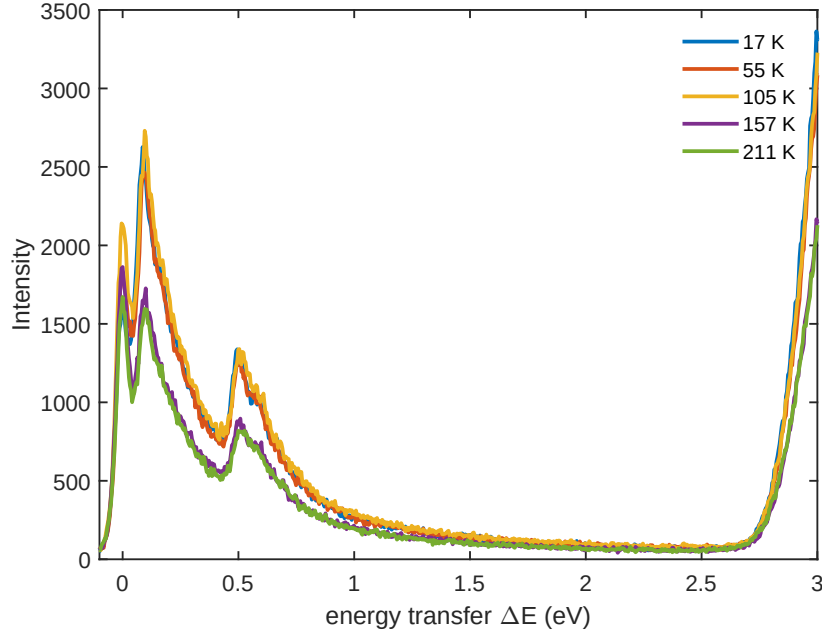
Supplementary Figure 8. Color-plot of intensity with the incident energy at the vertical axis and the energy loss on the horizontal axis. The narrow features around 0 – 0.5 eV of energy transfer are independent of the incident energy, representing localized *d-d* excitations. Strong *d-p* hybridization allows to observe excitations with a predominant *d*-character. The fluorescence appears above 3 eV of energy transfer for incident energies  $E_i > 527$  eV.

The comparison between energy transfer scans for different incident energies is shown in Supplementary Figure 9.

In all scans the features below 1 eV are well observed, without any apparent dependence on the incident energy value.



Supplementary Figure 9. Oxygen RIXS line-shape dependence on incident energy.

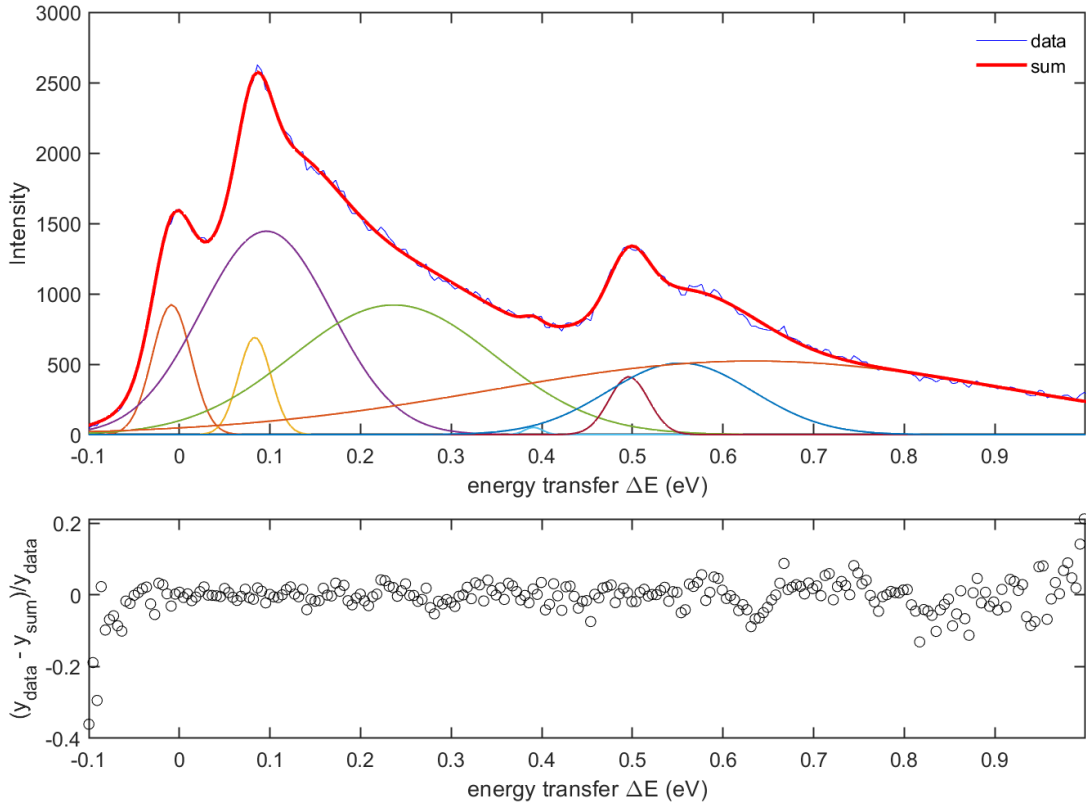


Supplementary Figure 10. Temperature dependence of the Oxygen *K* spectra.

In Supplementary Figure 10 we show energy transfer scans at  $E_{in} = 527.6$  eV for various temperatures with improved statistics. Alongside the sharp features characterizing the elastic line, the Jahn-Teller and the spin-orbit energy gaps, there are weak signs of additional features appearing above those  $d-d$  excitations. To analyze in detail the O *K*-edge line-shape we start with the assumption that all excitations can be described as Gaussians. Also, we focus on excitations at  $\Delta E < 1$  eV since above 1 eV there is a long, low-intensity tail, which is followed by a strong increase due to the fluorescence.

#### A. 8 independent Gaussian modes

We start with data obtained at  $T = 17$  K. Due to a highly asymmetric profile, a large number of modes is required. With 8 modes the overall goodness-of-fit is satisfactory but several modes have unreasonably large widths.



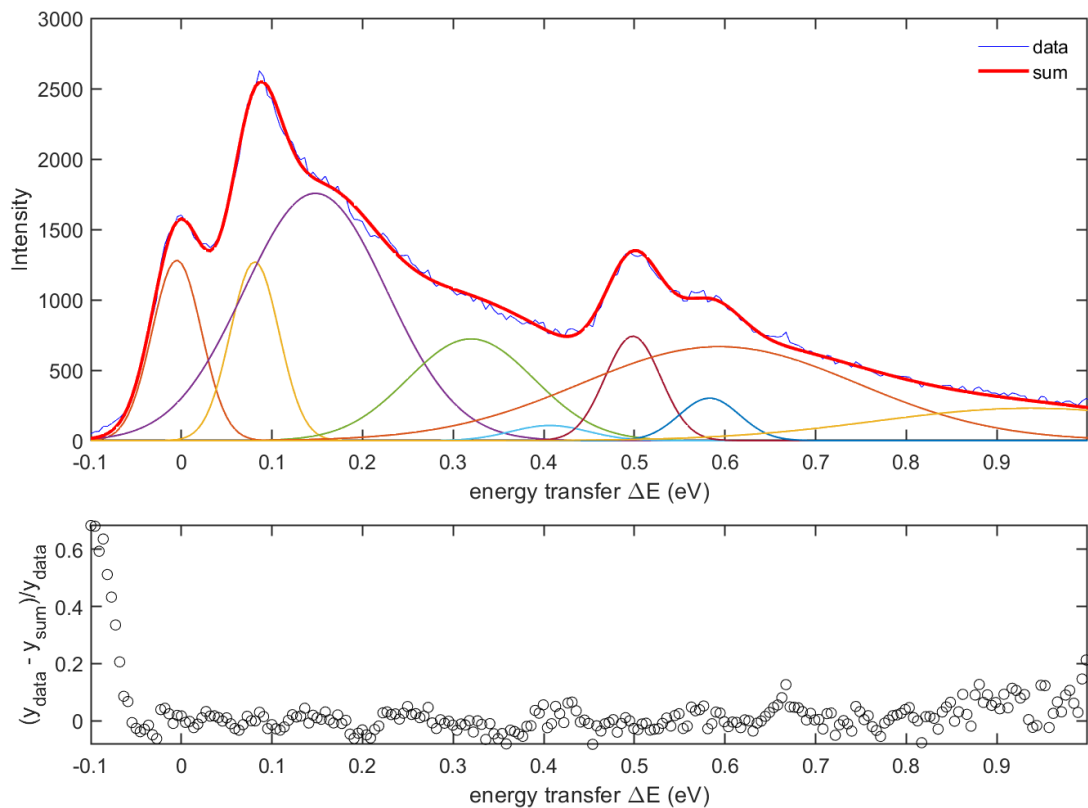
Supplementary Figure 11. Oxygen RIXS line-shape described with 8 Gaussian-type modes.

### B. 9 independent Gaussian modes

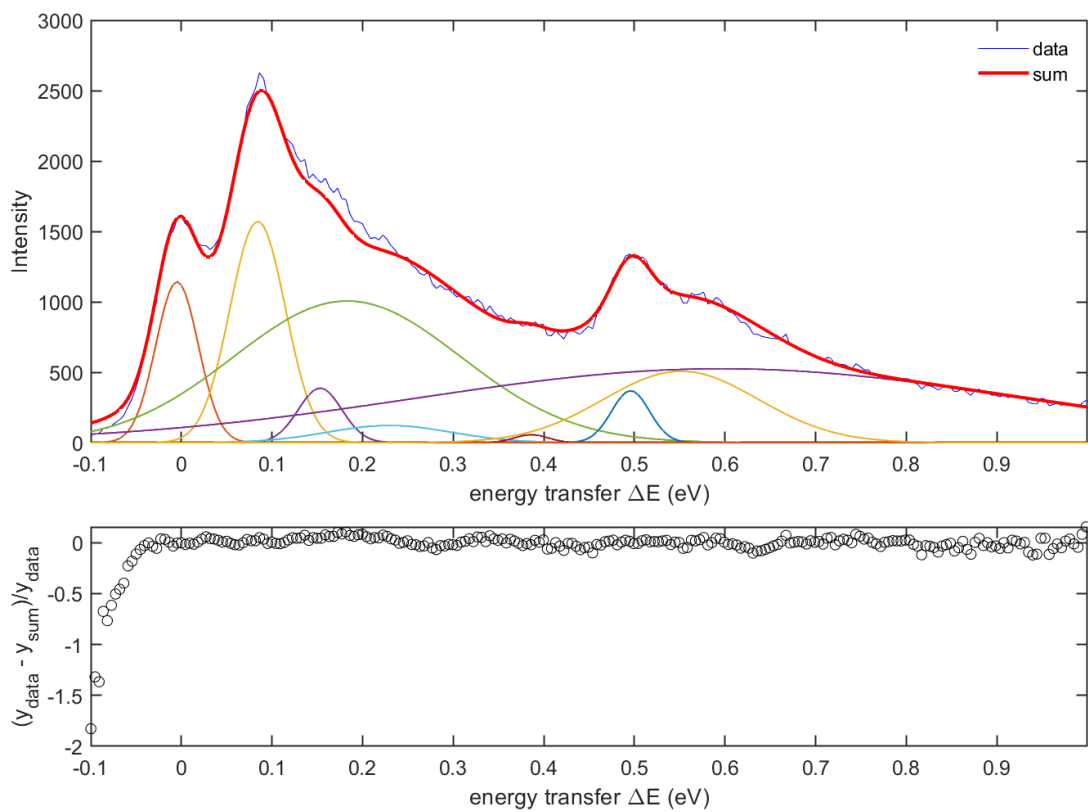
With 9 modes the agreement is slightly better, especially the behavior around the features that are  $\sim 100$  meV above  $d-d$  excitations.

### C. 10 independent Gaussian modes

Increasing the number of modes to 10 leads to a deterioration of the goodness-of-fit, with one mode being practically zero everywhere. Although we have tried to find an optimum set of initial guesses for the parameters of the fit (30 in total), we did not find that the fit converges to a better solution. It is obvious that with 30 parameters (or more if needed) the convergence to the global minimum becomes increasingly difficult.



Supplementary Figure 12. Oxygen RIXS line-shape described with 9 Gaussian-type modes.



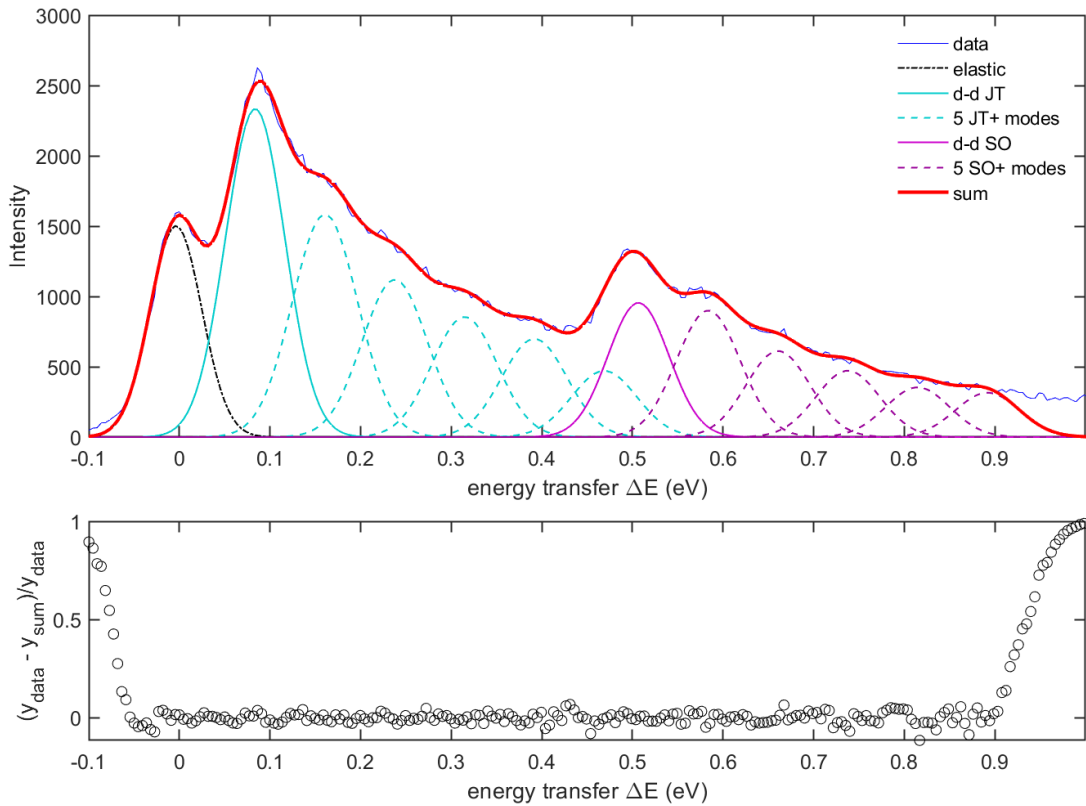
Supplementary Figure 13. Oxygen RIXS line-shape described with 10 Gaussian-type modes.

### D. 3 independent Gaussian modes + two sets of 5 dependent modes

Motivated by faint features in the RIXS line-shape that seem to be separated by a constant value (also seen in oxygen RIXS spectra of  $\text{Ba}_2\text{NaOsO}_6$  [S1]), we adopted the approach where the parameters describing the elastic line, together with two  $d-d$  excitations (we mark them JT and SO, for  $\sim 80$  meV and  $\sim 500$  meV features respectively) remain unrestrained, but other modes are grouped into sets. One set consists of excitations that are related to the JT excitation (JT+ modes), while the other set is related to the SO excitation (SO+ modes). The energy of the  $n$ -th mode is given by:

$$E_n = E_{d-d} + n * E_{mode} \quad (1)$$

the width is the same for all the mods within a set, while the amplitudes are free parameters. Within this approach we can describe the measured profile to a very good degree, using 5 additional modes in each set (13 modes in total), with significant deviations occurring at negative energy transfers as well as above 0.9 eV. In this case we are using 22 free parameters for the fit.



Supplementary Figure 14. Oxygen RIXS line-shape described with 13 Gaussian-type modes. The extracted values are listed in Table 2.

The resulting values are:

| mode       | elastic | JT          | 1     | 2     | 3     | 4     | 5     | SO           | 1     | 2     | 3     | 4     | 5     |
|------------|---------|-------------|-------|-------|-------|-------|-------|--------------|-------|-------|-------|-------|-------|
| Amplitude  | 1502    | 2335        | 1583  | 1119  | 854   | 698   | 471   | 955          | 900   | 612   | 472   | 354   | 315   |
| E (meV)    | -3.8    | <b>83.7</b> | +1*77 | +2*77 | +3*77 | +4*77 | +5*77 | <b>506.7</b> | +1*77 | +2*77 | +3*77 | +4*77 | +5*77 |
| FWHM (meV) | 67.2    | 77.2        | 85.1  | 85.1  | 85.1  | 85.1  | 85.1  | 79.5         | 82.9  | 82.9  | 82.9  | 82.9  | 82.9  |

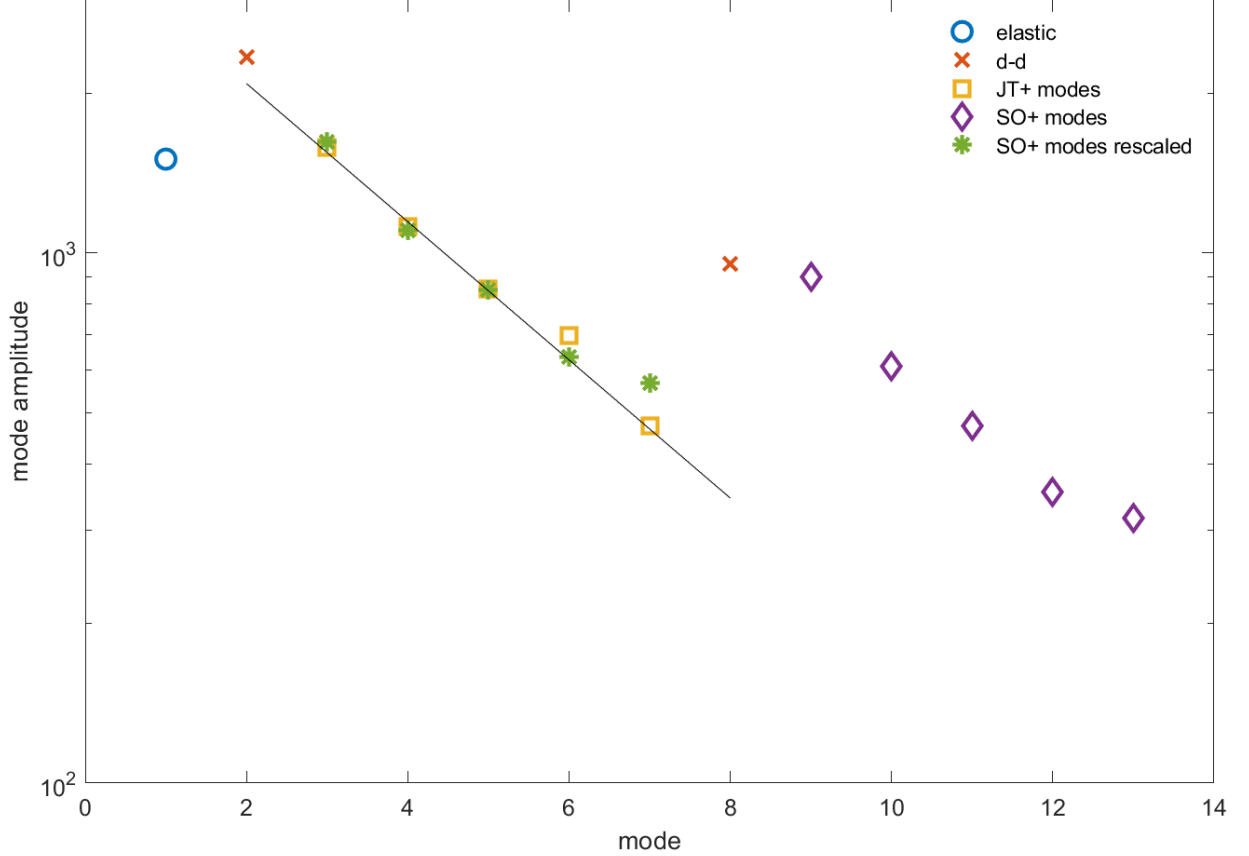
Supplementary Table 2. Extracted values of independent parameters for 13 Gaussian modes.

### E. 3 independent Gaussian modes + two sets of 5 dependent modes with an exponential decay of amplitude

In an attempt to further reduce the number of free parameters, we look at the amplitude dependence of modes. It can be concluded that the amplitudes of modes follow to a very good degree an exponential decay of the form:

$$A_n = A_1 \cdot e^{-n*B} \quad (2)$$

where  $A_1$  is the amplitude for  $n = 1$ . This leaves two  $A_1$  values and the decay rate  $B$  as independent parameters.



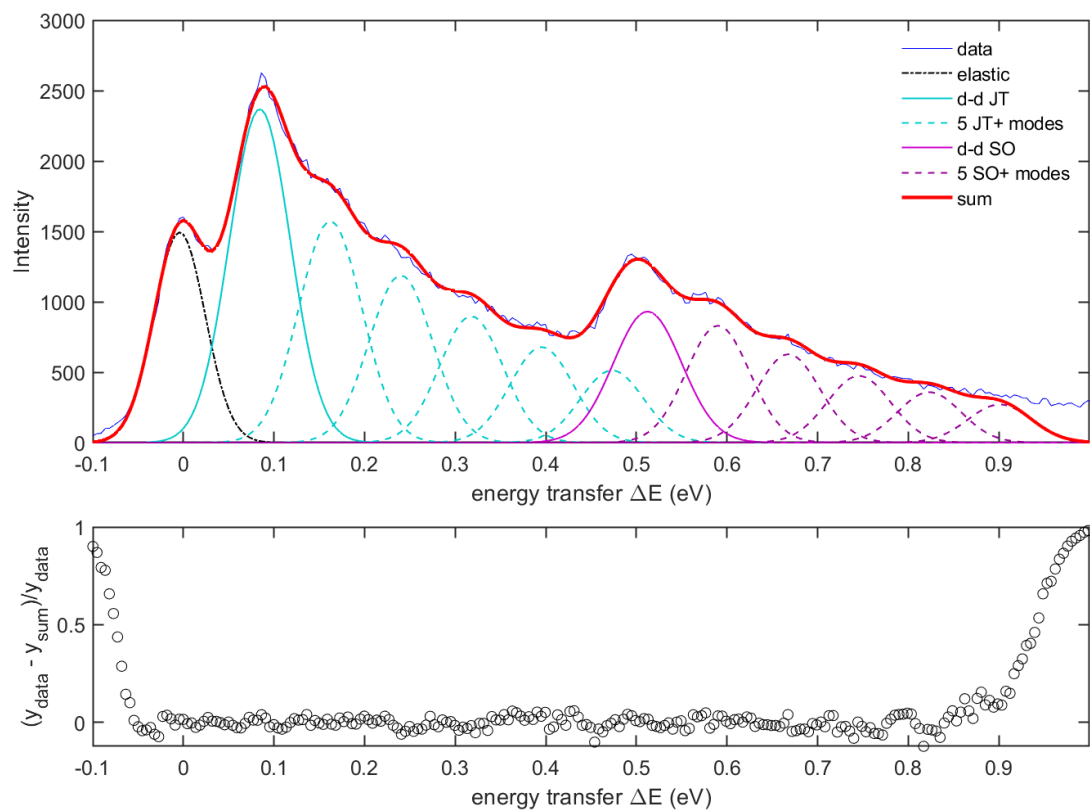
Supplementary Figure 15. Amplitudes of 13 modes in a semi-log scale. The solid line represents an exponential decay of amplitude with the mode number.

Additionally, the widths of modes for JT+ and SO+ sets are found to be very similar, therefore we set them to be equal. This way, the number of independent parameters decreases to 14.

The resulting values are:

| mode       | elastic | JT          | 1       | 2       | 3       | 4       | 5       | SO           | 1       | 2       | 3       | 4       | 5       |
|------------|---------|-------------|---------|---------|---------|---------|---------|--------------|---------|---------|---------|---------|---------|
| Amplitude  | 1493    | 2369        | 1572    | 1188    | 898     | 679     | 513     | 931          | 830     | 628     | 474     | 358     | 271     |
| E (meV)    | -4.1    | <b>84.5</b> | +1*77.8 | +2*77.8 | +3*77.8 | +4*77.8 | +5*77.8 | <b>512.3</b> | +1*77.8 | +2*77.8 | +3*77.8 | +4*77.8 | +5*77.8 |
| FWHM (meV) | 66.6    | 79.1        | 82.3    | 82.3    | 82.3    | 82.3    | 82.3    | 88.9         | 82.3    | 82.3    | 82.3    | 82.3    | 82.3    |

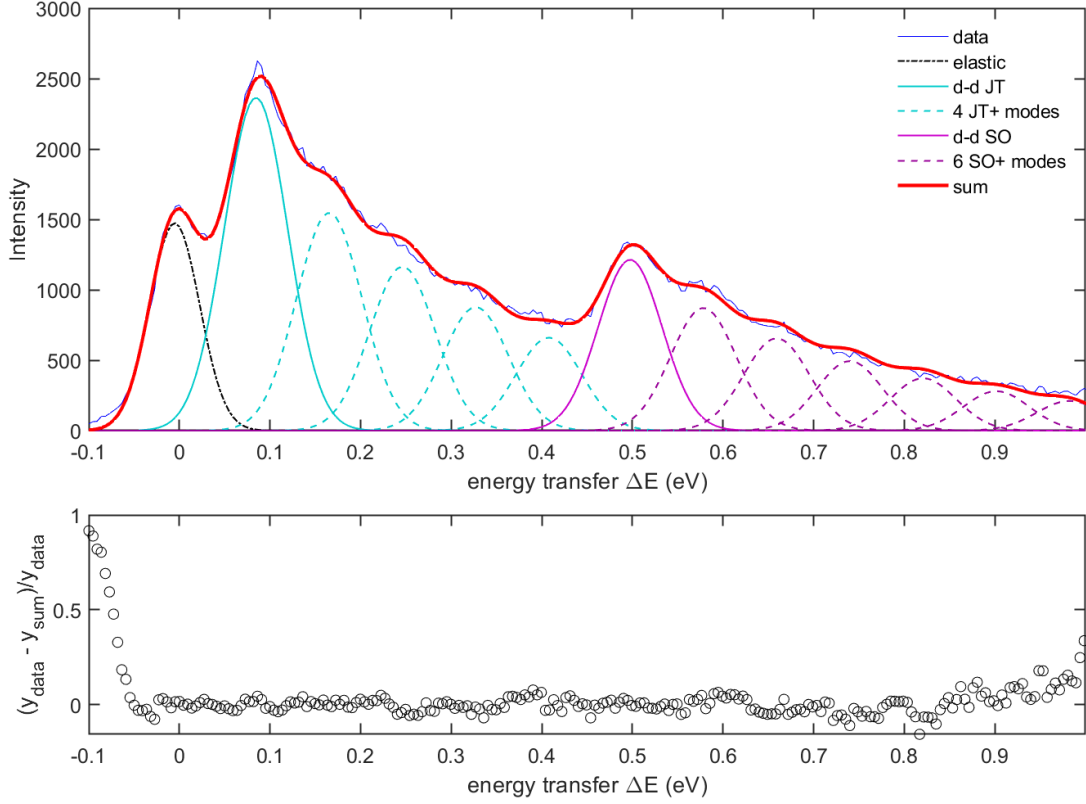
Supplementary Table 3. Extracted values of independent parameters for 13 Gaussian modes.



Supplementary Figure 16. Oxygen RIXS line-shape described with 13 Gaussian-type modes. The extracted values are listed in Table 3.

### F. 3 independent Gaussian modes + (4+6) dependent modes with an exponential decay of amplitude

As is evident from Figure 16, the 5th JT+ mode is strongly overlapping with the SO  $d-d$  excitation, indicating that the number of JT+ modes could be reduced to four. At the same time, to cover as much the high-energy side, we increase the number of SO+ modes to six.



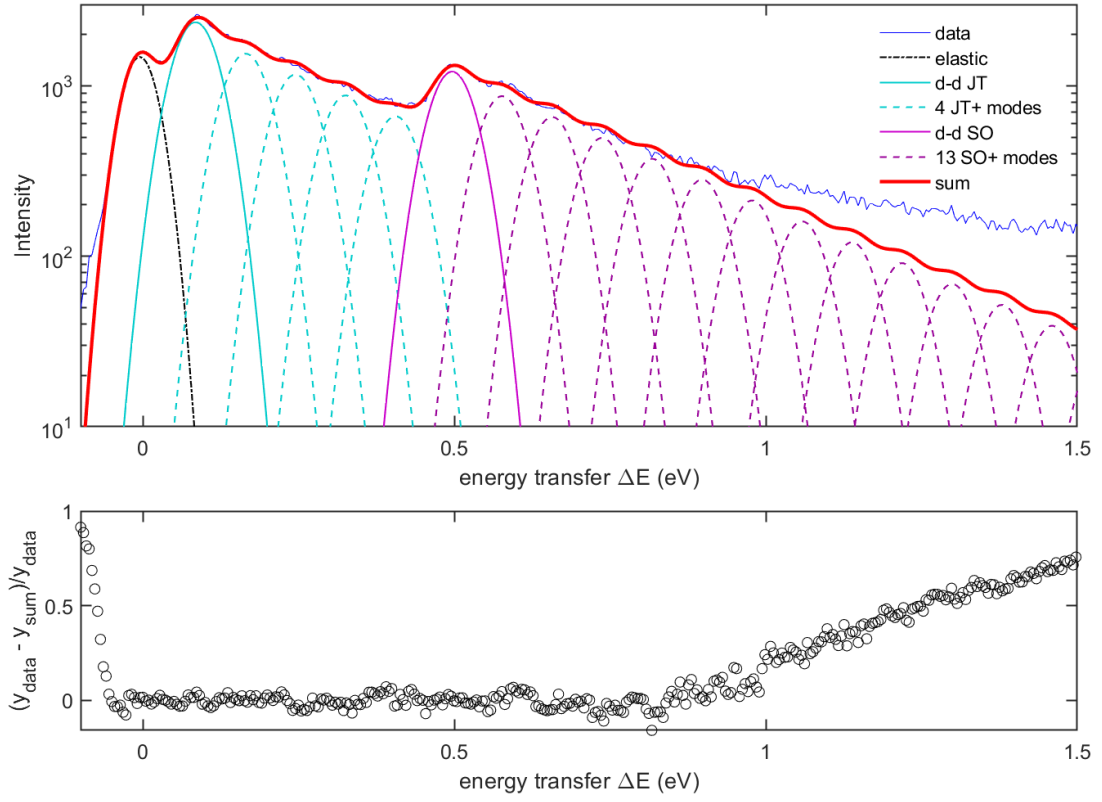
Supplementary Figure 17. Oxygen RIXS line-shape described with 13 Gaussian-type modes. The extracted values are listed in Table 4.

| mode       | elastic | JT          | 1       | 2       | 3       | 4       | SO           | 1       | 2       | 3       | 4       | 5       | 6       |
|------------|---------|-------------|---------|---------|---------|---------|--------------|---------|---------|---------|---------|---------|---------|
| Amplitude  | 1474    | 2365        | 1547    | 1165    | 877     | 660     | 1214         | 871     | 656     | 494     | 372     | 280     | 211     |
| E (meV)    | -5.1    | <b>82.5</b> | +1*80.8 | +2*80.8 | +3*80.8 | +4*80.8 | <b>497.5</b> | +1*80.8 | +2*80.8 | +3*80.8 | +4*80.8 | +5*80.8 | +6*80.8 |
| FWHM (meV) | 65.0    | 79.1        | 85.3    | 85.3    | 85.3    | 85.3    | 83.1         | 85.3    | 85.3    | 85.3    | 85.3    | 85.3    | 85.3    |

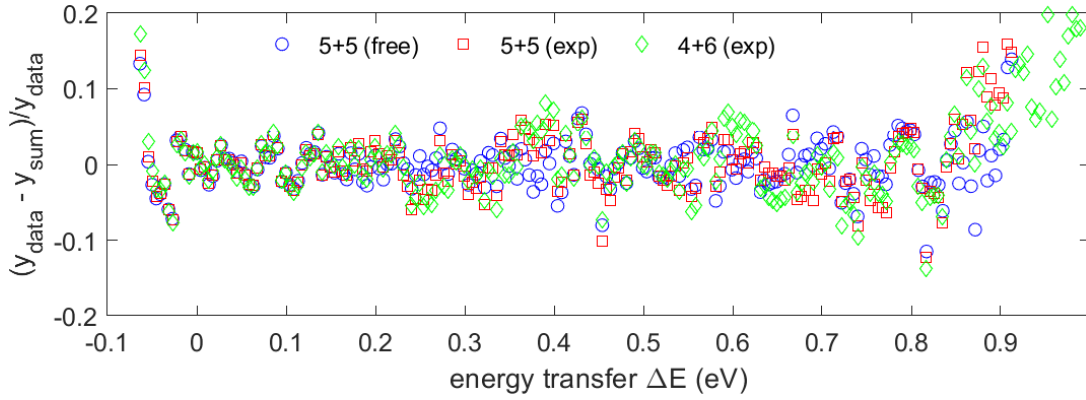
Supplementary Table 4. Extracted values of independent parameters for 13 Gaussian modes.

Increasing the number of SO+ modes further, in an attempt to cover the energy transfer range above 1 eV, results in a deterioration of the fit. The line-shape transitions into a long, low-intensity tail, which above 2.5 eV transforms into a fluorescence signal. No obvious features are present in the range 1 - 2.5 eV, and therefore we do not attempt to model it.

Finally, we summarize in one plot the relative errors obtained using three different approaches. No significant difference is noticed across the investigated range below 1 eV.



Supplementary Figure 18. Oxygen RIXS line-shape above 1 eV described with additional SO+ modes.

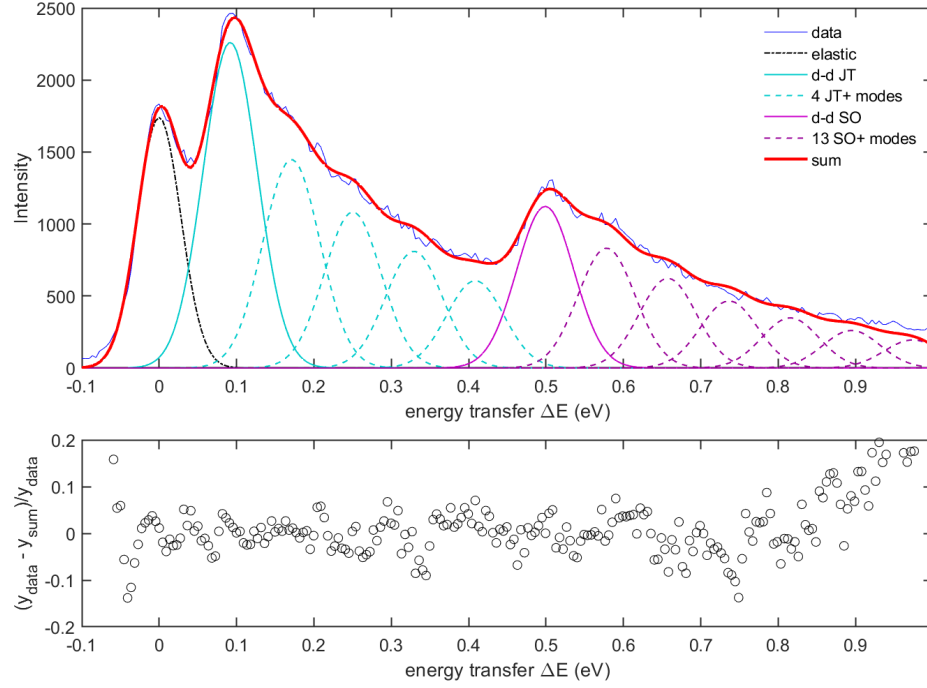


Supplementary Figure 19. Relative errors using three different approaches elaborated in subsections *D* (blue circles), *E* (red squares) and *F* (green diamonds). Two numbers for each approach indicate the total number of JT+ and SO+ modes used for the fit. 'free' indicates that the amplitudes were independent parameters, while 'exp' indicates that amplitudes follow the exponential decay.

### G. Temperature dependence of the oxygen RIXS line-shape

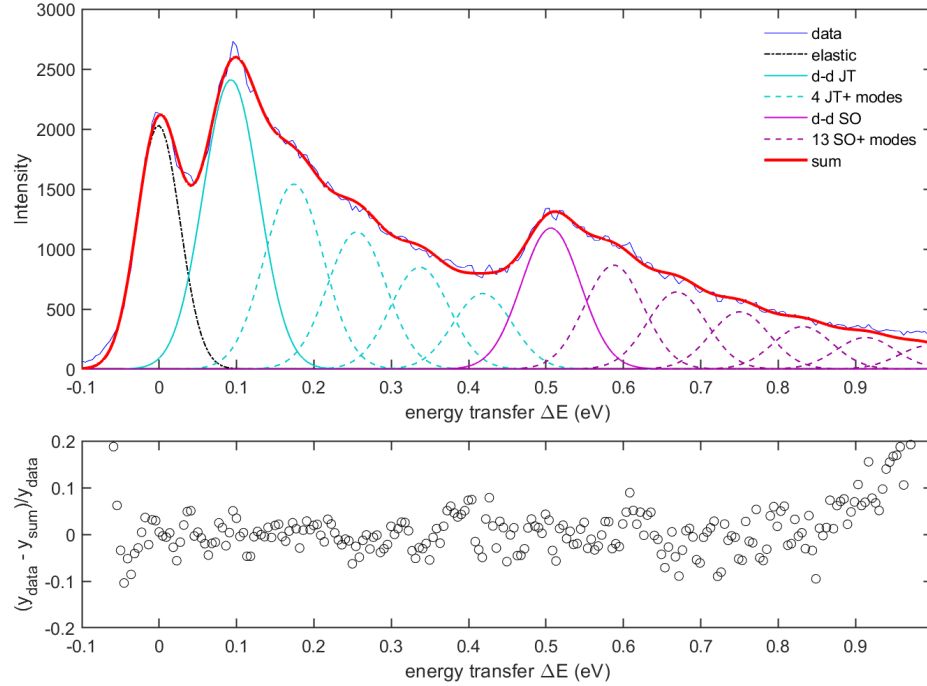
We use the approach elaborated in subsection *F* (4+6 dependent modes) to reveal the temperature evolution of the modes.

**T = 55 K**



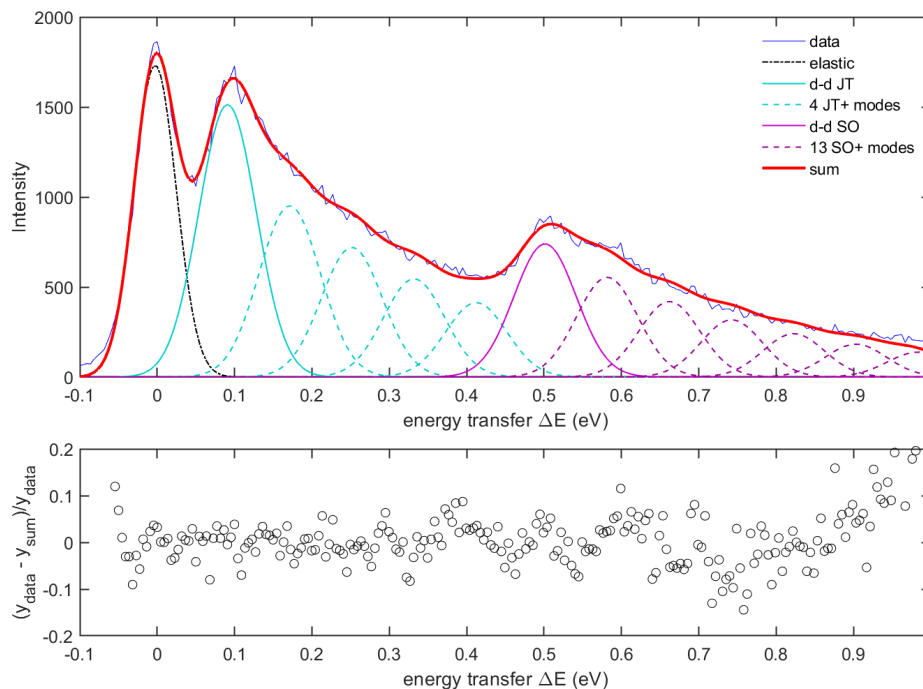
Supplementary Figure 20. Oxygen RIXS line-shape at 55 K.

**T = 105 K**



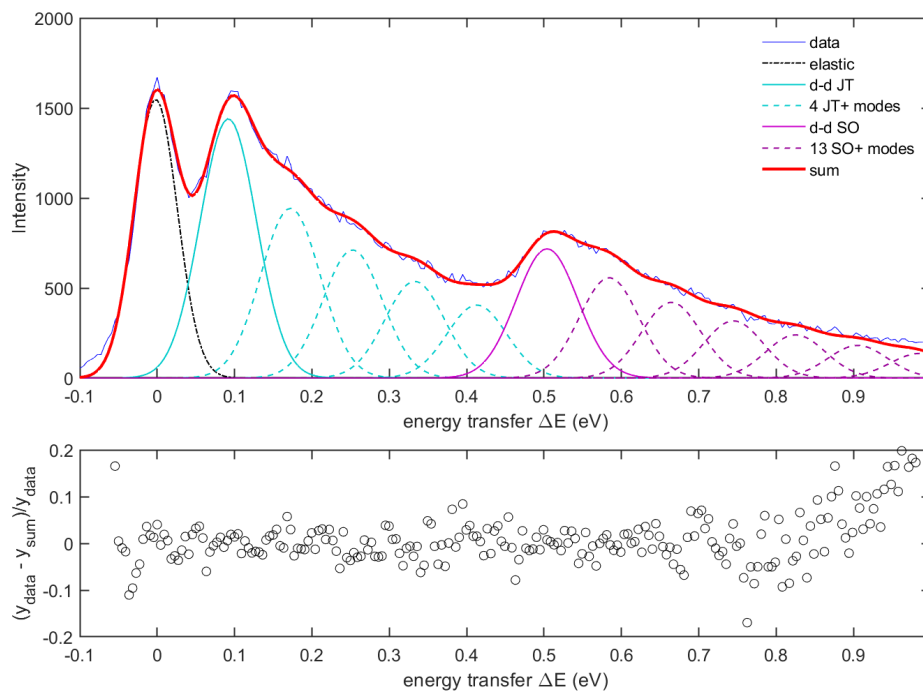
Supplementary Figure 21. Oxygen RIXS line-shape at 105 K.

**T = 157 K**



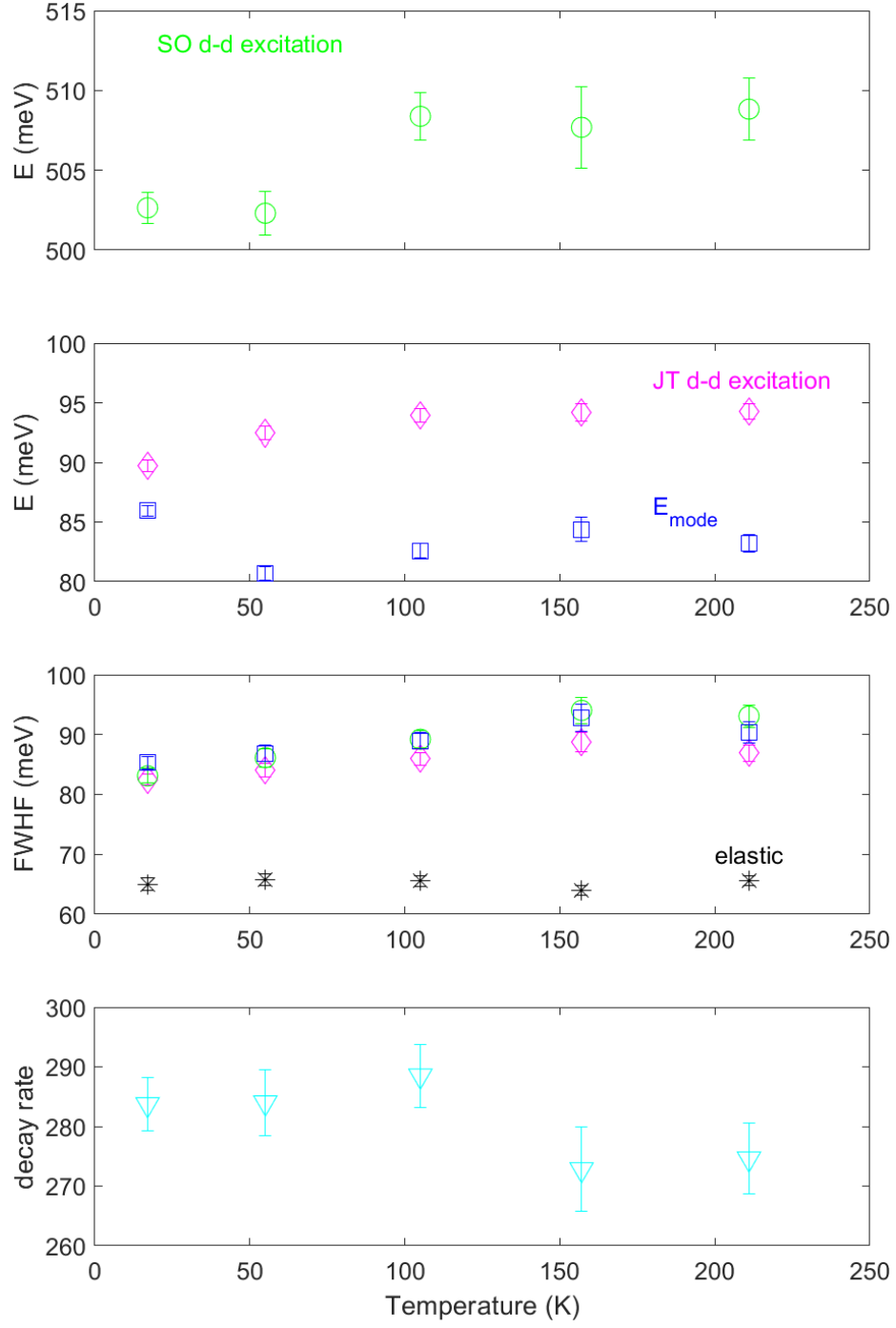
Supplementary Figure 22. Oxygen RIXS line-shape at 157 K.

**T = 211 K**



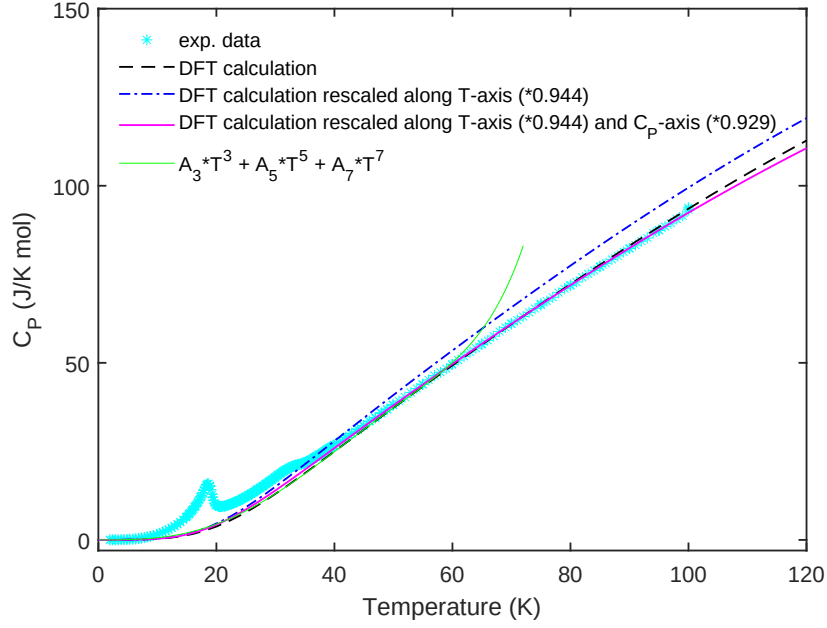
Supplementary Figure 23. Oxygen RIXS line-shape at 211 K.

The temperature dependence of extracted parameters is displayed in Figure 24. Very little variation is observed, confirming the hypothesis that the system does not change significantly when cooled down below  $T_Q$  and  $T_M$ .



Supplementary Figure 24. Temperature dependence of extracted parameters. The error bars are given as one standard deviation obtained from the fit.

## Supplementary Note 3. SPECIFIC HEAT



Supplementary Figure 25. The calculated values of phonon specific heat cross the experimentally obtained values, so the subtraction would result in negative values of electronic specific heat. By allowing the values to be renormalized along both axes, the segment between 60 K and 100 K is well reproduced. The rescaling factors are 0.944 along the temperature axis and 0.929 along the specific heat axis. We also plot a 7th order polynomial ( $A_3 = 6.3 \times 10^{-4}$ ;  $A_5 = -1.8 \times 10^{-7}$ ;  $A_7 = 2.0 \times 10^{-11}$ ) which at temperatures 50 - 60 K matches well with the experimental data. In all cases there is only a minor difference below 60 K, which does not affect the conclusion that the total entropy amounts to  $R \ln 2$ .

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- [S1] S. Agrestini, F. Borgatti, P. Florio, J. Frassinetti, D. Fiore Mosca, Q. Faure, B. Detlefs, C. J. Sahle, S. Francoual, J. Choi, M. Garcia-Fernandez, K.-J. Zhou, V. F. Mitrović, P. M. Woodward, G. Ghiringhelli, C. Franchini, F. Boscherini, S. Sanna, and M. Moretti Sala, Origin of magnetism in a supposedly nonmagnetic osmium oxide, *Phys. Rev. Lett.* **133**, 066501 (2024).