# The identity of the exchangeable sulfur-containing ligand at the Mo(V) center of R160Q human sulfite oxidase.

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## Supporting Information

**Generation of the phosphate form of R160Q from Species 1.**

The preparation and spectroscopy of Species 1 prepared at pH = ~6.4 is described in the manuscript. Species 1 was also prepared in $^{17}$O-enriched buffer at pH = ~5.4. The ESE detected spectrum of this sample (Figure S1) is identical in appearance to the previously reported EPR spectrum.[1] Phosphate buffer, which was prepared by dissolving natural abundance phosphate in $^{17}$O-enriched water (>90 atom % Aldrich 609862), was then incrementally added to this sample so that the final phosphate concentration was about 700 mM. This concentration is about 10 times greater than that used by Bray, *et al.*, to completely transform the low pH form (*lpH*) of chicken SO (cSO) into the phosphate-inhibited form ($P_i$).[2] The field sweep ESE-detected spectrum of the sample obtained in the presence of 700 mM phosphate is also presented in Figure S1. Analysis of the initial and final spectra revealed the generation of a new enzyme form. The *g*-values of this new form (1.952, 1.968, 2.00) are different from those of $P_i$ (1.965, 1.973, 1.995). Unfortunately, the inefficient conversion of Species 1 into this new form (~20%) and the overlap of the EPR spectra of these samples complicated our attempts to quantitatively investigate the substitution of the sulfur-containing ligand by phosphate. To overcome these problems, it was necessary to investigate the samples at a slightly higher pH (~6.4, as described in the manuscript). The resulting spectra of the phosphate forms prepared at pH = 5.4 and 6.4 are nearly identical (Figure S2).

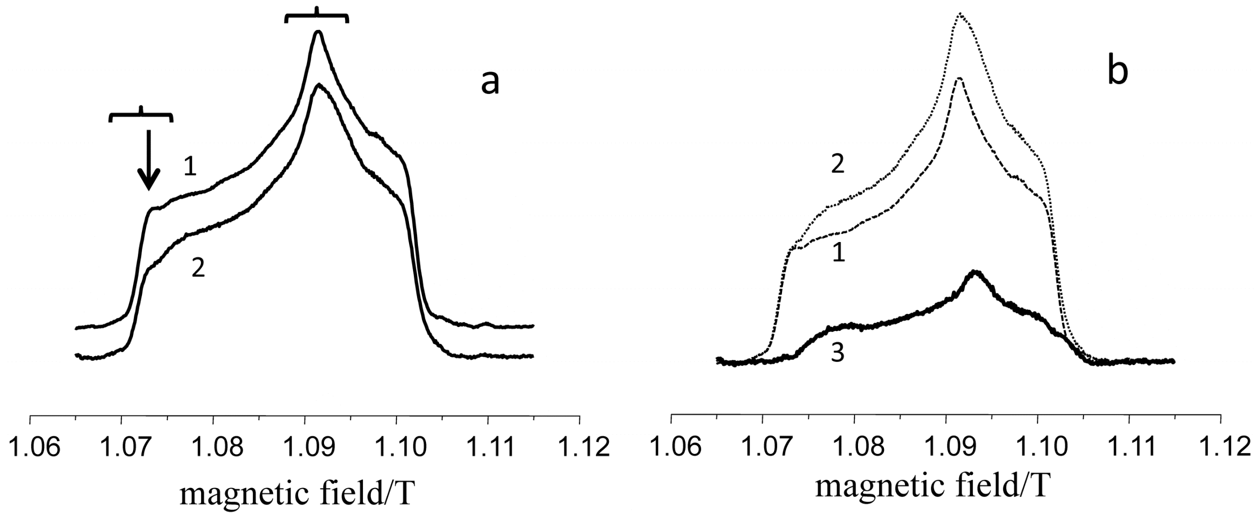


**Figure S1**. Transformation of the ESE-detected EPR spectrum of Species 1 by addition of phosphate. Panel "a" Traces 1 and 2 are the initial spectrum and the spectrum obtained in the presence of 700 mM phosphate, respectively. Brackets show the field regions in the spectra where the effects of phosphate are clearest. The arrow indicates the low-field shoulder position of Species 1 at which the intensity of trace 1 was adjusted for subtraction from trace 2, as shown in panel "b". Panel "b", trace 3 shows the EPR spectrum of the new form of R160Q generated by phosphate addition. This was obtained by subtracting trace 1 from 2, where trace 2 is the same as trace 2 in panel "a" and trace 1 is the amplitude-adjusted spectrum of Species 1. Experimental conditions: $\nu_{mw}$ = 30.216 GHz; Temperature = 20K; two-pulse ESE pulse durations = 20 ns.

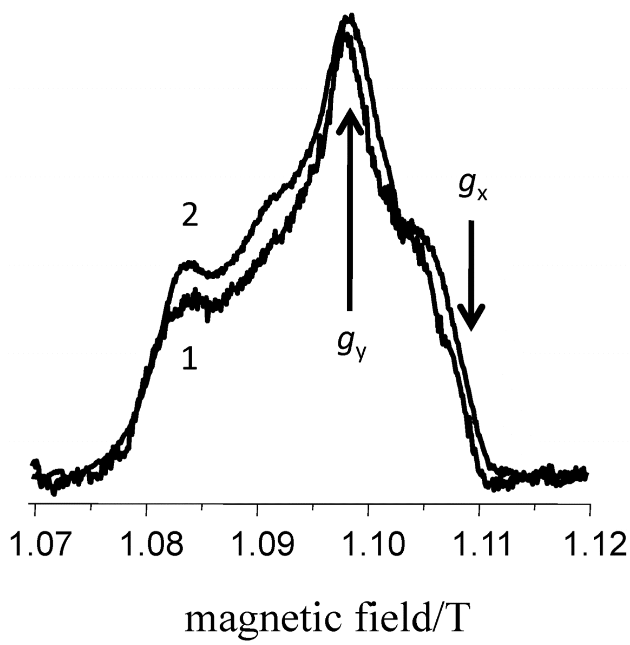


**Figure S2**. The EPR spectra of the new form obtained by addition of phosphate to Species 1. Trace 1 is the same as trace 3 of Figure S1, panel "b". Trace 2 is the spectrum presented in Figure 2 in the manuscript. These are shown together here for comparison. Trace 2 has been shifted by ~40 G to offset the difference of mw frequencies at which these spectra were recorded.

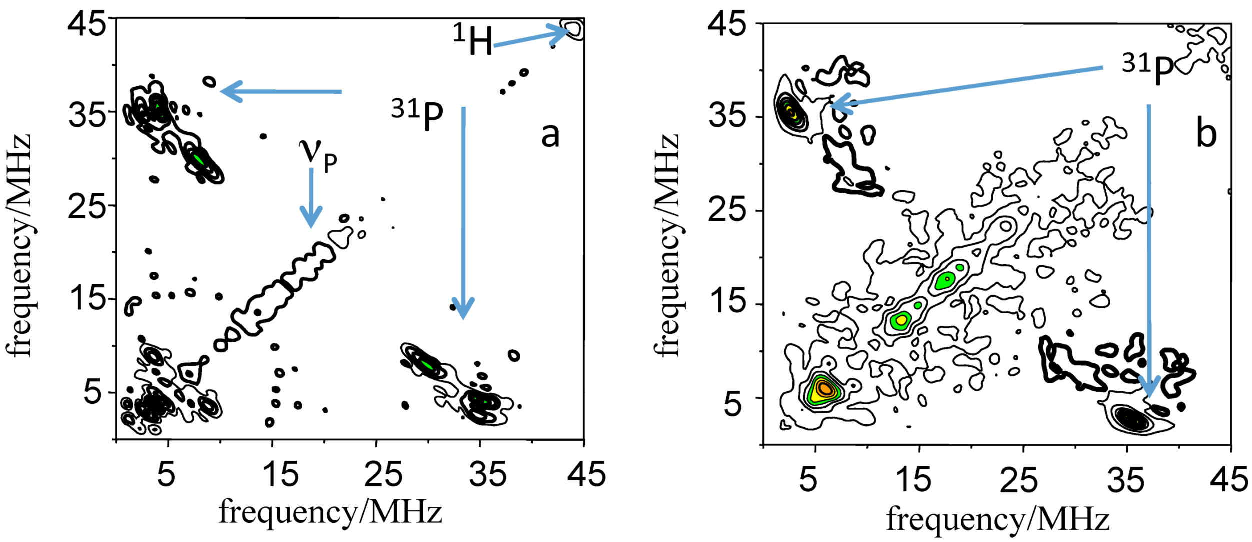


**Figure S3**. $K_a$-band HYSCORE spectra of Species 1P obtained at the $g_y$ (panel "a") and $g_x$ (panel "b") EPR positions (indicated by the arrows in Figure S2). Experimental conditions: mw frequency = 30.216 GHz; Temperature = 20K; mw pulses = 12, 12, 22, and 12 ns. The spectra represent the sum of the spectra obtained at time intervals between the first two mw pulses of 150, 180, and 210 ns.

**Preparation of the standard *lpH* form of R160Q by $CaCl_2$ addition.**

An additional experimental effort was made to chemically distinguish either sulfate or sulfite as the identity of the sulfur-containing ligand of Species 1. The rational of this approach is described in the manuscript. Two cations ($Ag^+$ and $Ca^{2+}$) were considered for these experiments based on the solubilities of their sulfate and sulfite salts in water. Although the $K_{sp}$ values of $Ag_2SO_3$ and $Ag_2SO_4$ differ by ten orders of magnitude,[3] this metal was eliminated as a candidate for our initial attempts (with limited enzyme available) since its redox activity could lead to ambiguous results or other complications ($Ag^0$, $Ag^+$, and $Ag^{2+}$ are potentially accessible). $Ca^{2+}$ is redox inert in water, and the $K_{sp}$ values of $CaSO_4$ and $CaSO_3$ ($9.1 \times 10^{-6}$ M and $6.8 \times 10^{-8}$ M, respectively) still differ sufficiently for our purposes.

For these experiments it is necessary to maintain a rather low pH (ideally pH $\leq$ 6) to prevent any transformation of Species 1 into Species 2. Simultaneously, it is also necessary that the pH be high enough (preferably pH $\geq$ 7) to avoid shifting the equilibrium of $SO_3^{2-}$ toward $HSO_3^-$ (a very soluble product). The optimal conditions for carrying out such experiments most effectively, although possible to find, would require large amounts of enzyme to identify the effects of multiple independent variables (including buffer pH, enzyme concentration, cation concentration, anion concentration, and total ion strength) on each other and on the results. Due to our limited amount of purified enzyme, however, we were only able to investigate the effects of buffered *vs.* unbuffered $CaCl_2$ solutions of various concentrations. In one of these trials, a substantial change in the initial EPR spectrum was observed. In that case, the sample was prepared at an initial pH of 5.8. This was then titrated by an unbuffered, 200 mM aqueous $CaCl_2$ solution (at pH = ~9) to a final $CaCl_2$ concentration of about 50 mM. The initial and final CW-EPR spectra are shown in Figure S4. The initial spectrum is primarily from Species 1, but Species 2 is also present in a very small amount. The addition of $CaCl_2$ resulted in the unexpected generation of a new species that was not Species 2. Since Species 1 remains in significant quantities, however, the amplitude-adjusted initial spectrum was subtracted from the final spectrum to isolate the EPR spectrum of this the new form (Figure S4, panel “b”). To adjust the amplitude of the initial spectrum, the unobstructed and clearly visible high-field signal at $g_x$ (marked by an arrow) was used. This kind of a spectrum has never been observed for R160Q previously. The splitting at the $g_z$ position (~9 G) is characteristic for EPR spectra of standard *lpH* forms, however, and is due to proton hyperfine coupling from coordinated hydroxo.[4, 5] For comparison, the CW-EPR spectrum of the *lpH* form of cSO is presented in Figure S4, panel “c”.

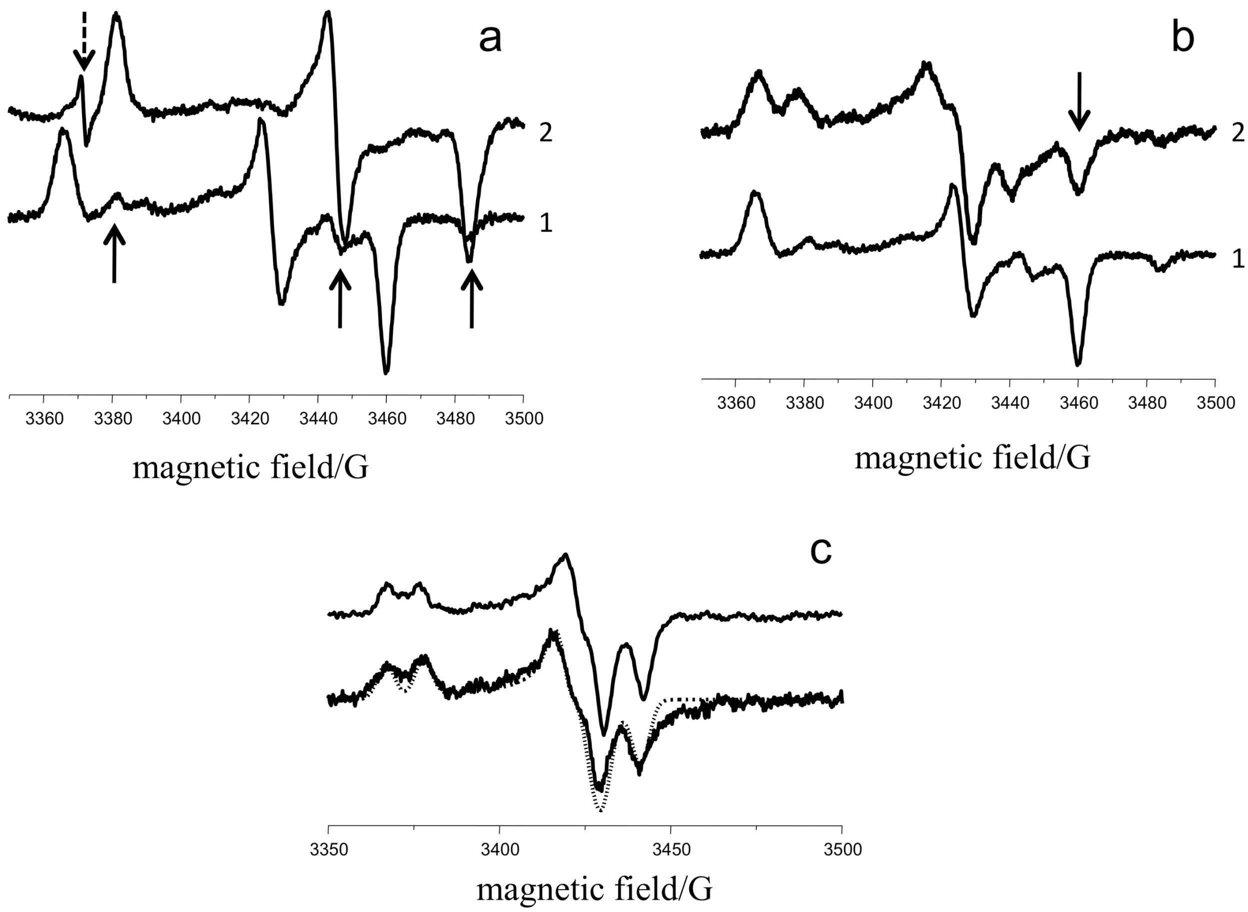


**Figure S4**. (a) The CW-EPR spectra of the initial R160Q sample (trace 1) and Species 2 (trace 2) obtained in separate preparations. The arrows at bottom spectrum point to the spectral features of Species 2 that are present in the initial spectrum in detectable (but negligible) amounts. The dashed arrow indicates the position of the DPPH *g*-marker ($g = 2.0036$). (b) The CW-EPR spectra recorded before (trace 1) and after (trace 2) the titration with $CaCl_2$. The arrow indicates the part of the spectra that was matched in amplitude for subtraction. As is evident from trace 2, the titration produced no additional amount of Species 2. (c) The upper trace is the CW-EPR spectrum of the *lpH* form of cSO. The lower trace (solid line) is the resulting difference spectrum from panel “b”. The dotted line shows the simulation of this trace (simulation parameters: $g = \{2.004; 1.974; 1.966\}$; $\mathbf{T} = \{5; 7; -12\}$ MHz; $a_{iso} = 25$ MHz). Experimental conditions: all spectra are adjusted to a mw frequency of 9.454 MHz; Temperature = 77K.

The magnetic resonance parameters that describe the spectrum of the new enzyme form are derived from the simulation of the spectrum. These are $g$ = {2.004; 1.974; 1.966}, T = {5; 7; -12} MHz, and $a_{iso}$ = 25 MHz. These parameters are very close to those of *lpH* cSO, which are $g$ = {2.0037, 1.972, 1.9658}, $a_{iso}$ = 26 MHz, and $\mathbf{T}_{\perp}$ = 5,[4, 5] and to the parameters of *lpH* forms from other sources.[6] These parameters provide clear evidence that Species 1 is transformed to a standard *lpH* form by $CaCl_2$. Although the details of this transformation are not yet understood, no other methods to produce the *lpH* form of R160Q are known. For example, the one-electron reduction of R160Q at low pH (from its Mo(VI) resting state) by Ti(III) generates only Species 2, while the same procedure for the *blocked* form of *Arabidopsis thaliana* SO (At-SO) generates the standard *lpH* form.[7] Also, the presence of high $Cl^-$ concentrations (~500 mM), which normally transforms other *blocked* forms to the *lpH* form, has no effect on Species 1.[7] One possible explanation for the conversion of Species 1 to the *lpH* form by $CaCl_2$ is the removal of the sulfite ligand from Mo(V) (and the solution) through the formation of $CaSO_3$. In this case, the open coordination site of Mo(V) could become accessible to water (or hydroxide). Alternatively, it is also possible that $CaCl_2$ could initially increase the solution pH sufficiently to transform Species 1 into Species 2, which may then release the sulfur-containing ligand, ultimately forming $CaSO_3$. To determine which of these (or other) possibilities is correct, or what the specific effect(s) on the active site structure may be, would be far beyond the scope of this paper.

**Spectral simulations**

Since the set of experimental spectra is incomplete (*i.e.* there is a lack of data at the $g_z$ position), there are many possible combinations of *hfi* and *nqi* parameters that could be used to simulate them. To simulate the spectra at the $g_y$ and $g_x$ positions collected at $K_a$- and W-bands, certain constrains must be imposed. As we have learned from our numerous previous simulations, the magnitudes of the isotropic and anisotropic *hfi* must be close to estimates made from the direct analysis of the experimental data, despite the possible broad variation of the *hfi* tensor axis within the *g*-frame. For example, Figure S5 shows a comparison of the experimental W- and $K_a$-band spectra (black lines) with one of the simulations (colored lines). For this particular simulation, we set $a_{iso}$ to -9 MHz and the extremely rhombic tensor of the anisotropic *hfi* to $\mathbf{T}$ = {1; 9; -10}. The elements of this tensor correlate with elements of the axial *hfi* tensor for which $T_{||}$ = -9 MHz, as evaluated in the manuscript.

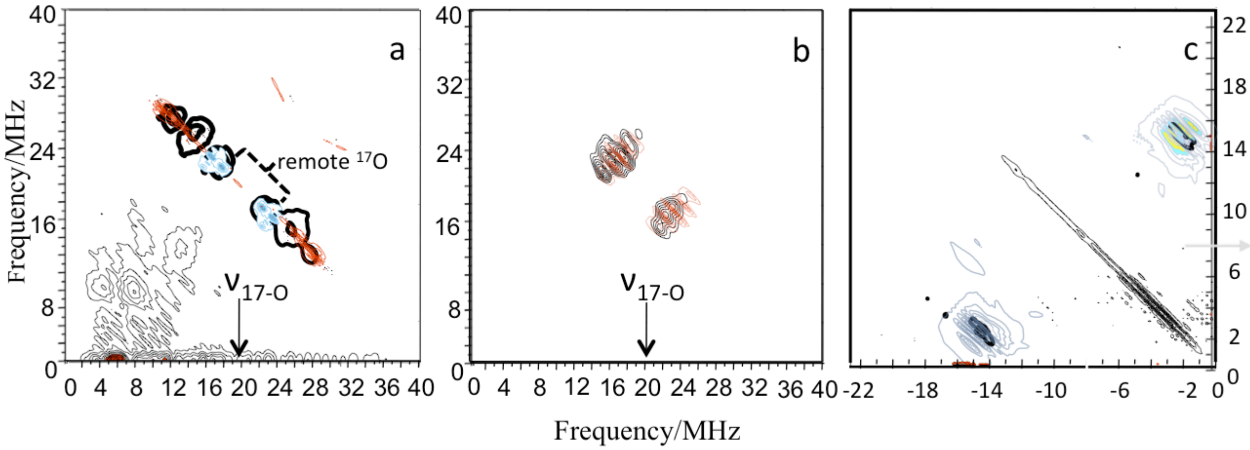


**Figure S5**. Experimental (black) and simulated (colored) W-(panels a and b) and $K_a$-band (panel c) HYSCORE contour plots of Species 1. (a) and (b) were collected at the $g_y$ and $g_x$ positions, indicated by the arrows in Figure S6. The simulated contours of the Mo(V)-coordinated $^{17}O$ (of the sulfite ligand) are shown as red lines in (a) and (b), and the simulated contours of the remote $^{17}O$ nuclei of sulfite are shown in blue in (a). Simulation parameters for the Mo(V)-coordinated $^{17}O$: $a_{iso}$ = -9 MHz; $a_{iso}$ distribution width = 3 MHz; $T_{11}$= 1 MHz; $T_{22}$ = -9 MHz; $\vartheta(hfi)$ = 90°; $\varphi(hfi)$ = 90°;$\psi(hfi)$ = 60°; $e^2Qq/h$ = 6.8 MHz; $\eta$ = 0.5; $\vartheta$ $(nqi)$ = 90°; $\varphi(nqi)$ = 0°; $\psi(nqi)$ = 90°. Simulation parameters for the remote $^{17}O$ nuclei of sulfite: $a_{iso}$ = 5.5 MHz; $a_{iso}$ distribution width = 1 MHz; $T_{11}$ = -1 MHz; $T_{22}$ = -1 MHz; $\vartheta(hfi)$ = 10°; $\varphi(hfi)$ = 5°; $\psi(hfi)$ = 0°; $e^2Qq/h$ = 4.8 MHz; $\eta$ = 0.5; $\vartheta(nqi)$ = 5°; $\varphi(nqi)$ = 0°; $\psi(nqi)$ = 0°. All of the simulations were performed using the SimBud software,[8] to which we refer for the definitions of all of the above parameters.

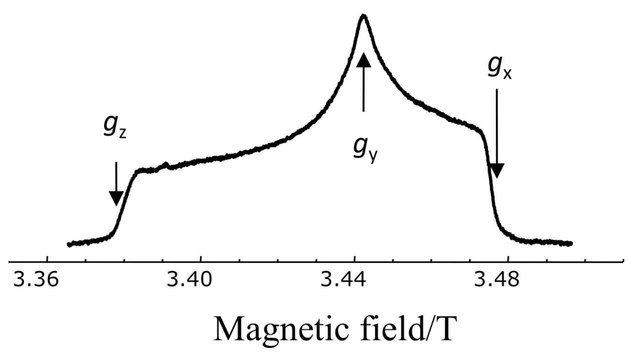


**Figure S6**. The W-band, two-pulse ESE field sweep spectrum of Species 1. The arrows show the approximate positions of $g_x$, $g_y$, and $g_z$ (1.9513, 1.9701, and 2.0062, respectively). Experimental conditions: microwave frequency = 94.89675 GHz; Temperature = 20K; pulse durations = 25 ns; pH = 5.8.

**Optimized sulfate- and sulfite-bound model coordinates and DFT-calculated magnetic resonance parameters.**

The DFT model preparations and property calculations were performed as described in the Materials and Methods section using the ORCA computational package.[9] The calculated magnetic resonance parameters for the sulfate-coordinated model (model 1) and one of the sulfite-coordinated models (model 2) are provided in

Tables 1 and S1, and the magnetic resonance parameters for all of the models are provided in Table S1 (below). For comparison, the parameters of the relevant enzymes are also provided in each table.

(1) SO-$SO_4$ (259 atoms):

| | | | |
|---|---|---|---|
| C | 27.974530 | 101.424337 | 27.378348 |
| C | 29.141937 | 101.527265 | 26.391898 |
| O | 29.073711 | 101.013873 | 25.266692 |
| C | 27.558518 | 99.967311 | 27.617460 |
| C | 28.553699 | 99.111392 | 28.411863 |
| C | 29.844431 | 98.730669 | 27.668299 |
| N | 30.453083 | 97.538948 | 28.272931 |
| C | 31.533626 | 97.476022 | 29.050897 |
| N | 32.314634 | 98.544053 | 29.302419 |
| N | 31.875754 | 96.277858 | 29.585959 |
| N | 30.273890 | 102.167159 | 26.821088 |
| C | 31.497505 | 102.062592 | 26.048733 |
| C | 32.449345 | 101.022432 | 26.631186 |
| O | 32.595890 | 100.916240 | 27.869868 |
| N | 33.113864 | 100.290789 | 25.723976 |
| C | 34.233072 | 99.425179 | 26.067949 |
| C | 34.524103 | 98.378680 | 24.967906 |
| C | 33.736177 | 97.108373 | 25.038806 |
| N | 32.384033 | 96.952739 | 24.734742 |
| C | 34.170307 | 95.857751 | 25.410394 |
| C | 32.028224 | 95.664708 | 24.923604 |
| N | 33.102682 | 94.986363 | 25.332838 |
| C | 22.904770 | 95.470018 | 27.085096 |
| C | 22.590919 | 94.131442 | 27.749159 |
| O | 21.621773 | 93.989525 | 28.497891 |
| C | 23.506303 | 96.482063 | 28.064685 |
| S | 25.217925 | 96.092257 | 28.694797 |
| N | 23.474892 | 93.117951 | 27.486610 |
| C | 23.291081 | 91.785670 | 28.053404 |
| C | 23.505107 | 91.716125 | 29.572179 |
| O | 23.054335 | 90.768424 | 30.224363 |
| C | 24.170428 | 90.769371 | 27.321204 |
| N | 24.229799 | 92.719149 | 30.127711 |
| C | 24.308299 | 92.845661 | 31.566890 |
| C | 23.520941 | 92.186405 | 36.721380 |
| C | 24.858921 | 92.575611 | 36.068912 |
| C | 25.978056 | 91.586263 | 36.416035 |
| C | 27.298044 | 91.832504 | 35.678185 |
| N | 27.218716 | 91.499215 | 34.245520 |
| C | 27.466189 | 92.294895 | 33.184367 |
| N | 27.646458 | 93.612337 | 33.311819 |
| N | 27.560439 | 91.737046 | 31.961753 |
| C | 31.090589 | 92.720596 | 34.904552 |
| C | 30.578736 | 93.998415 | 35.550737 |
| O | 30.451659 | 94.118766 | 36.771247 |
| N | 30.226821 | 95.013793 | 34.695306 |
| C | 29.646688 | 96.246109 | 35.221358 |
| C | 28.416107 | 95.936294 | 36.066844 |
| O | 27.526575 | 95.133756 | 35.702045 |
| C | 29.328082 | 97.256408 | 34.108580 |
| C | 30.561989 | 97.916242 | 33.456187 |
| C | 30.097800 | 98.790603 | 32.283111 |
| C | 31.388419 | 98.741497 | 34.453691 |

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| C | 27.286534 | 96.454139 | 38.194853 |
| C | 25.985249 | 97.231727 | 37.910950 |
| O | 25.543788 | 98.060776 | 38.706831 |
| N | 25.382778 | 96.898776 | 36.740596 |
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| C | 23.727347 | 97.077735 | 34.907511 |
| C | 24.689515 | 97.333705 | 33.779672 |
| C | 25.428576 | 96.385243 | 33.102360 |
| C | 24.996972 | 98.593973 | 33.144707 |
| N | 26.179873 | 96.977797 | 32.101491 |
| C | 25.920899 | 98.334060 | 32.093864 |
| C | 24.552509 | 99.914625 | 33.357209 |
| C | 26.390539 | 99.353975 | 31.259061 |
| C | 25.025907 | 100.931424 | 32.533341 |
| C | 25.937475 | 100.651247 | 31.492407 |
| C | 31.877676 | 87.150217 | 25.704645 |
| C | 30.437532 | 87.069702 | 25.187981 |
| O | 30.168729 | 86.663420 | 24.051270 |
| C | 32.451696 | 88.578152 | 25.793482 |
| C | 32.081824 | 89.392654 | 27.039114 |
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| O | 32.563507 | 90.551418 | 27.182707 |
| N | 29.488180 | 87.475248 | 26.081621 |
| C | 28.089753 | 87.626807 | 25.698886 |
| C | 27.839191 | 88.766107 | 24.678487 |
| C | 28.470573 | 90.068769 | 25.110038 |
| C | 28.123315 | 90.660410 | 26.332563 |
| C | 29.483865 | 90.683766 | 24.359237 |
| C | 28.779782 | 91.790274 | 26.815601 |
| C | 30.149603 | 91.822724 | 24.821949 |
| C | 29.817761 | 92.362295 | 26.069364 |
| O | 30.530882 | 93.453734 | 26.525320 |
| C | 29.433303 | 86.918821 | 33.757420 |
| C | 29.569456 | 86.899429 | 32.225629 |
| C | 30.975414 | 86.540362 | 31.714590 |
| C | 31.966911 | 87.715415 | 31.694708 |
| N | 31.605267 | 88.716437 | 30.691080 |
| C | 31.168292 | 89.967415 | 30.895740 |
| N | 31.173059 | 90.540963 | 32.130746 |
| N | 30.661861 | 90.633893 | 29.854149 |
| C | 24.390154 | 88.371937 | 31.888017 |
| C | 25.070034 | 88.114603 | 30.525204 |
| C | 26.266852 | 89.043354 | 30.311610 |
| C | 25.475630 | 86.641651 | 30.373803 |
| S | 28.520878 | 94.513268 | 29.632424 |
| O | 27.601273 | 93.324168 | 29.592074 |
| O | 28.396041 | 95.318981 | 30.912284 |
| O | 30.008771 | 94.084106 | 29.428043 |
| O | 28.258101 | 95.459663 | 28.383699 |
| N | 30.279469 | 99.423025 | 19.648968 |
| C | 31.433105 | 100.045923 | 19.717443 |
| N | 32.003113 | 100.543295 | 18.575238 |
| N | 32.126671 | 100.173424 | 20.890404 |
| C | 31.674560 | 99.636298 | 22.117054 |
| O | 32.420348 | 99.770840 | 23.122753 |
| N | 29.847808 | 98.435053 | 23.248855 |
| C | 28.772036 | 97.442940 | 22.979695 |
| C | 27.826995 | 97.866517 | 21.830276 |
| N | 28.587087 | 98.255415 | 20.682324 |

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| C | 29.776397 | 98.911609 | 20.808619 |
| C | 27.914308 | 97.195065 | 24.197508 |
| S | 28.588948 | 96.161782 | 25.477143 |
| C | 26.659107 | 97.691197 | 24.293038 |
| S | 25.637818 | 97.308040 | 25.697790 |
| C | 26.033654 | 98.575799 | 23.225756 |
| O | 27.016558 | 98.968387 | 22.245473 |
| C | 25.419475 | 99.866468 | 23.757001 |
| Mo | 26.694191 | 95.525906 | 26.885319 |
| O | 33.622853 | 92.418761 | 25.708740 |
| O | 31.538260 | 93.415601 | 31.564420 |
| O | 26.058468 | 94.084633 | 26.194096 |
| H | 27.131298 | 101.972001 | 26.929777 |
| H | 28.214745 | 101.924979 | 28.331283 |
| H | 26.603349 | 99.969670 | 28.162947 |
| H | 27.355525 | 99.489022 | 26.648061 |
| H | 28.820375 | 99.603588 | 29.361650 |
| H | 28.040257 | 98.172993 | 28.676545 |
| H | 29.615613 | 98.474545 | 26.624300 |
| H | 30.572491 | 99.548098 | 27.668414 |
| H | 29.850996 | 96.703210 | 28.236196 |
| H | 33.183615 | 98.386091 | 29.800694 |
| H | 32.282410 | 99.431191 | 28.763516 |
| H | 31.200230 | 95.495562 | 29.535716 |
| H | 32.503352 | 96.272336 | 30.382215 |
| H | 30.347518 | 102.452183 | 27.794120 |
| H | 31.214886 | 101.819916 | 25.016516 |
| H | 32.026508 | 103.029794 | 26.047248 |
| H | 32.826671 | 100.329032 | 24.730789 |
| H | 35.135699 | 100.044398 | 26.205373 |
| H | 34.034237 | 98.933078 | 27.030030 |
| H | 34.398148 | 98.846245 | 23.979626 |
| H | 35.582295 | 98.093973 | 25.053647 |
| H | 31.733637 | 97.669326 | 24.357272 |
| H | 35.154557 | 95.524540 | 25.714582 |
| H | 31.022934 | 95.265580 | 24.826132 |
| H | 33.170380 | 93.922372 | 25.519372 |
| H | 21.942044 | 95.876510 | 26.737605 |
| H | 23.555536 | 95.352278 | 26.207092 |
| H | 23.567435 | 97.477351 | 27.603054 |
| H | 22.886910 | 96.558126 | 28.969268 |
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| H | 25.233664 | 91.037369 | 27.404899 |
| H | 24.501615 | 93.514266 | 29.548261 |
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| H | 25.021967 | 93.639503 | 31.812879 |
| H | 23.328479 | 93.099672 | 32.003189 |
| H | 23.608695 | 92.148561 | 37.817773 |
| H | 22.730439 | 92.909235 | 36.475946 |
| H | 23.185184 | 91.196552 | 36.377578 |
| H | 24.730019 | 92.614705 | 34.974000 |
| H | 25.158611 | 93.586589 | 36.387307 |
| H | 26.192452 | 91.631714 | 37.496596 |
| H | 25.639058 | 90.552742 | 36.221655 |
| H | 27.601585 | 92.881047 | 35.784651 |
| H | 28.098075 | 91.218365 | 36.122521 |

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| H | 27.603284 | 94.117127 | 34.218139 |
| H | 27.895289 | 94.163524 | 32.481074 |
| H | 27.336433 | 90.754443 | 31.829397 |
| H | 27.508196 | 92.324381 | 31.111203 |
| H | 31.951526 | 92.360148 | 35.481953 |
| H | 30.303362 | 91.955451 | 34.965574 |
| H | 31.370782 | 92.870094 | 33.855029 |
| H | 30.315057 | 94.888194 | 33.689662 |
| H | 30.392031 | 96.692339 | 35.902690 |
| H | 28.712013 | 96.768338 | 33.336856 |
| H | 28.695903 | 98.050268 | 34.539601 |
| H | 31.210568 | 97.115695 | 33.053101 |
| H | 29.553604 | 98.193185 | 31.537392 |
| H | 29.417858 | 99.583046 | 32.632345 |
| H | 30.950714 | 99.274971 | 31.785136 |
| H | 31.814861 | 98.130301 | 35.262747 |
| H | 32.230606 | 99.235833 | 33.946889 |
| H | 30.771530 | 99.531386 | 34.911583 |
| H | 29.123792 | 97.228135 | 37.458646 |
| H | 27.065217 | 95.377558 | 38.263563 |
| H | 27.658554 | 96.797252 | 39.165845 |
| H | 25.841470 | 96.223830 | 36.123969 |
| H | 23.366491 | 97.391908 | 37.028309 |
| H | 24.346395 | 98.658688 | 36.284943 |
| H | 23.515001 | 95.997259 | 34.964150 |
| H | 22.761696 | 97.562446 | 34.688026 |
| H | 25.469028 | 95.311250 | 33.255042 |
| H | 26.762559 | 96.475120 | 31.430839 |
| H | 23.839103 | 100.139800 | 34.152564 |
| H | 27.076818 | 99.131533 | 30.445917 |
| H | 24.685273 | 101.956696 | 32.687114 |
| H | 26.285069 | 101.465227 | 30.853082 |
| H | 32.477379 | 86.563731 | 24.996193 |
| H | 31.951219 | 86.669966 | 26.692197 |
| H | 33.553229 | 88.534948 | 25.768887 |
| H | 32.156925 | 89.171579 | 24.912768 |
| H | 29.841514 | 87.946558 | 26.921761 |
| H | 27.737139 | 86.678793 | 25.264691 |
| H | 27.518679 | 87.803580 | 26.621568 |
| H | 28.247261 | 88.455353 | 23.707031 |
| H | 26.748178 | 88.876654 | 24.554875 |
| H | 27.324638 | 90.220778 | 26.934723 |
| H | 29.772058 | 90.248763 | 23.398614 |
| H | 28.478080 | 92.222876 | 27.770820 |
| H | 30.960261 | 92.267408 | 24.242465 |
| H | 30.293910 | 93.619199 | 27.466462 |
| H | 29.673272 | 85.934127 | 34.185776 |
| H | 30.104126 | 87.656203 | 34.227546 |
| H | 28.405832 | 87.167779 | 34.060874 |
| H | 28.856369 | 86.163494 | 31.822556 |
| H | 29.261535 | 87.868248 | 31.798005 |
| H | 31.407634 | 85.735983 | 32.333015 |
| H | 30.910897 | 86.137609 | 30.691154 |
| H | 32.016801 | 88.185640 | 32.688054 |
| H | 32.981465 | 87.351069 | 31.469450 |
| H | 31.585503 | 88.438984 | 29.690992 |
| H | 31.781708 | 90.133657 | 32.832777 |
| H | 31.133640 | 91.568689 | 32.131070 |
| H | 30.802510 | 90.172677 | 28.919668 |

| | | | |
|---|---|---|---|
| H | 30.502858 | 91.636311 | 29.905340 |
| H | 23.534535 | 87.696490 | 32.033416 |
| H | 25.098202 | 88.188673 | 32.716952 |
| H | 24.010708 | 89.401044 | 31.940337 |
| H | 24.323400 | 88.352884 | 29.749767 |
| H | 27.033917 | 88.864807 | 31.090472 |
| H | 26.758618 | 88.868584 | 29.342790 |
| H | 25.948153 | 90.096063 | 30.337962 |
| H | 26.206083 | 86.352329 | 31.148251 |
| H | 24.605494 | 85.976853 | 30.477159 |
| H | 25.933623 | 86.443949 | 29.392444 |
| H | 31.390365 | 100.542241 | 17.764836 |
| H | 32.649191 | 101.323643 | 18.633986 |
| H | 33.052582 | 100.598616 | 20.920177 |
| H | 29.485855 | 99.179575 | 23.863086 |
| H | 29.255301 | 96.499307 | 22.672381 |
| H | 27.182721 | 97.009673 | 21.555892 |
| H | 28.117810 | 98.351016 | 19.784610 |
| H | 25.249297 | 97.980736 | 22.710813 |
| H | 24.611162 | 99.647554 | 24.466813 |
| H | 26.189319 | 100.468131 | 24.258704 |
| H | 25.004585 | 100.443756 | 22.919924 |
| H | 33.186422 | 91.673210 | 26.259555 |
| H | 34.113539 | 91.970319 | 25.000442 |
| H | 32.407973 | 93.305994 | 31.141119 |
| H | 30.925764 | 93.694152 | 30.822181 |

The *g*-tensor orientation is given by the following points:

| | | | |
|---|---|---|---|
| origin | 28.446148691 | 94.912523803 | 28.894120295 |
| gx | 28.150858310 | 95.594840763 | 28.616071159 |
| gy | 27.940262511 | 94.942548973 | 29.505055501 |
| gz | 28.981822966 | 95.317006460 | 29.317807699 |

Euler rotation of the hyperfine tensor to the *g*-tensor:

| Atom | Alpha | Beta [degrees] | Gamma | Ax | Ay [MHz] | Az |
|---|---|---|---|---|---|---|
| 122Mo | -21.9 | 60.5 | 35.2 | 142.77 | 47.98 | 51.00 |
| 125O | 0.2 | 68.4 | -11.4 | -0.45 | 0.92 | 9.30 |
| 102O | 4.8 | 62.8 | 20.4 | -3.50 | -20.78 | -4.39 |
| 98S | 16.1 | 69.9 | 9.6 | 2.94 | 2.81 | 4.26 |
| 100O | 29.4 | 54.5 | 31.2 | -0.06 | -0.04 | -3.58 |
| 99O | -2.7 | 51.7 | -78.4 | 0.03 | -0.34 | 0.38 |
| 101O | 38.4 | 39.1 | 47.7 | 0.01 | 0.29 | -0.26 |

Euler rotation of the Electric Field Gradient (EFG) tensor to the *g*-tensor:

| Atom | Alpha | Beta [degrees] | Gamma | EFGx | EFGy [a.u.**-3] | EFGz |
|---|---|---|---|---|---|---|
| 122Mo | -8.8 | 54.0 | 5.4 | 1.17 | -1.28 | 0.11 |
| 125O | -25.9 | 71.9 | -21.3 | -0.19 | -0.00 | 0.19 |
| 102O | 12.9 | 71.5 | 28.9 | -0.64 | -1.15 | 1.78 |
| 98S | -15.0 | 72.4 | 19.8 | -0.30 | -0.04 | 0.34 |
| 100O | 4.7 | 41.2 | -84.3 | 1.74 | -0.77 | -0.97 |
| 99O | 7.5 | 54.6 | 24.8 | 1.72 | -0.97 | -0.75 |
| 101O | 27.3 | 63.3 | -52.2 | -0.95 | -0.83 | 1.78 |

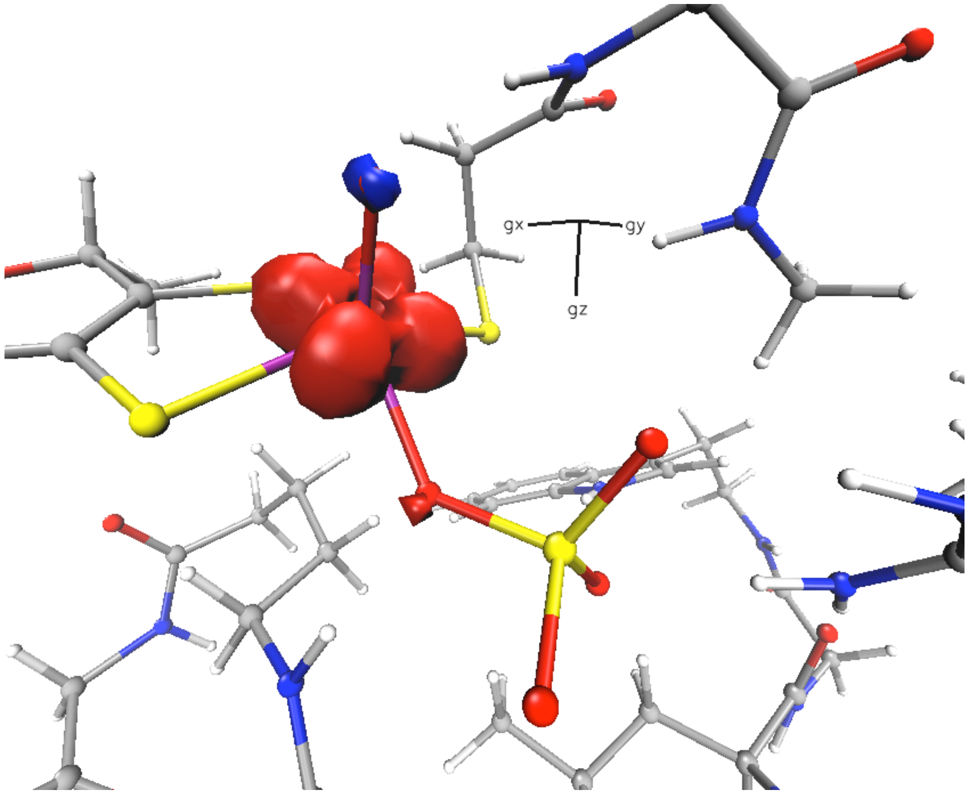


**Figure S7**. The *g*-tensor orientation and total spin density of **1** (contoured at 0.01 e/bohr$^3$).

(2) SO-$SO_3$ (258 atoms):

| | | | |
|---|---|---|---|
| C | 27.974511 | 101.423733 | 27.377982 |
| C | 29.142034 | 101.526724 | 26.391676 |
| O | 29.074677 | 101.011478 | 25.267366 |
| C | 27.519425 | 99.973551 | 27.585619 |
| C | 28.485110 | 99.080304 | 28.374345 |
| C | 29.773149 | 98.679297 | 27.637512 |
| N | 30.353594 | 97.480538 | 28.254518 |
| C | 31.417539 | 97.409365 | 29.051323 |
| N | 32.213673 | 98.467317 | 29.308243 |
| N | 31.728632 | 96.210487 | 29.604609 |
| N | 30.271809 | 102.173139 | 26.817845 |
| C | 31.497604 | 102.062223 | 26.048793 |
| C | 32.436345 | 101.007287 | 26.628866 |
| O | 32.566900 | 100.882507 | 27.866689 |
| N | 33.115253 | 100.290132 | 25.718818 |
| C | 34.233372 | 99.425022 | 26.068428 |
| C | 34.523231 | 98.365432 | 24.981067 |
| C | 33.722166 | 97.104127 | 25.060214 |
| N | 32.370514 | 96.965680 | 24.745967 |
| C | 34.134838 | 95.851328 | 25.452144 |
| C | 31.992261 | 95.686752 | 24.949054 |
| N | 33.052803 | 94.997196 | 25.378301 |
| C | 22.890665 | 95.474335 | 27.124814 |
| C | 22.593442 | 94.132577 | 27.786559 |
| O | 21.648566 | 93.983699 | 28.565437 |
| C | 23.515393 | 96.478670 | 28.098694 |
| S | 25.223878 | 96.055466 | 28.712393 |
| N | 23.466870 | 93.118286 | 27.490007 |

| | | | |
|---|---|---|---|
| C | 23.291721 | 91.784724 | 28.052721 |
| C | 23.553340 | 91.702767 | 29.563794 |
| O | 23.125462 | 90.748354 | 30.218061 |
| C | 24.128893 | 90.760715 | 27.281701 |
| N | 24.293633 | 92.705524 | 30.120249 |
| C | 24.308415 | 92.844893 | 31.566305 |
| C | 23.520459 | 92.185722 | 36.720715 |
| C | 24.858491 | 92.575013 | 36.068404 |
| C | 25.974794 | 91.575850 | 36.397648 |
| C | 27.297161 | 91.828858 | 35.664870 |
| N | 27.222158 | 91.511469 | 34.226992 |
| C | 27.421469 | 92.336662 | 33.182023 |
| N | 27.581338 | 93.650221 | 33.312870 |
| N | 27.498190 | 91.795533 | 31.939007 |
| C | 31.090295 | 92.720446 | 34.904825 |
| C | 30.568316 | 93.993876 | 35.551774 |
| O | 30.425931 | 94.108471 | 36.771568 |
| N | 30.224761 | 95.011627 | 34.696062 |
| C | 29.646081 | 96.245857 | 35.221349 |
| C | 28.418841 | 95.934802 | 36.070468 |
| O | 27.526810 | 95.133384 | 35.706762 |
| C | 29.314334 | 97.248062 | 34.104422 |
| C | 30.538080 | 97.906390 | 33.432041 |
| C | 30.053583 | 98.787445 | 32.272189 |
| C | 31.387243 | 98.724071 | 34.416621 |
| N | 28.336154 | 96.640717 | 37.208109 |
| C | 27.285537 | 96.453788 | 38.194541 |
| C | 25.981408 | 97.222052 | 37.899621 |
| O | 25.528641 | 98.043591 | 38.697259 |
| N | 25.385554 | 96.893855 | 36.723579 |
| C | 24.170532 | 97.570284 | 36.296731 |
| C | 23.675999 | 97.087584 | 34.926285 |
| C | 24.613928 | 97.348143 | 33.782975 |
| C | 25.331617 | 96.387670 | 33.100672 |
| C | 24.944511 | 98.610595 | 33.162734 |
| N | 26.091102 | 96.966016 | 32.101146 |
| C | 25.862205 | 98.326794 | 32.103254 |
| C | 24.552796 | 99.949728 | 33.368467 |
| C | 26.360063 | 99.323977 | 31.258675 |
| C | 25.063703 | 100.945942 | 32.539836 |
| C | 25.959308 | 100.637464 | 31.490841 |
| C | 31.878970 | 87.149868 | 25.705174 |
| C | 30.440747 | 87.080181 | 25.180485 |
| O | 30.174509 | 86.698264 | 24.035519 |
| C | 32.449934 | 88.577685 | 25.812819 |
| C | 32.065368 | 89.382485 | 27.061209 |
| O | 31.282186 | 88.831114 | 27.922392 |
| O | 32.533656 | 90.545957 | 27.209262 |
| N | 29.489096 | 87.469235 | 26.079372 |
| C | 28.091011 | 87.626165 | 25.698925 |
| C | 27.828999 | 88.807723 | 24.733016 |
| C | 28.456343 | 90.098492 | 25.205931 |
| C | 28.164684 | 90.623848 | 26.472320 |
| C | 29.413635 | 90.773963 | 24.433414 |
| C | 28.818675 | 91.750981 | 26.966440 |
| C | 30.072860 | 91.909855 | 24.908845 |
| C | 29.797974 | 92.391240 | 26.194315 |
| O | 30.504546 | 93.478447 | 26.653400 |
| C | 29.433601 | 86.918511 | 33.757648 |
| C | 29.569949 | 86.899086 | 32.225875 |

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|---|---|---|---|
| C | 30.982848 | 86.569195 | 31.715087 |
| C | 31.950950 | 87.763571 | 31.703583 |
| N | 31.578596 | 88.755969 | 30.695490 |
| C | 31.172547 | 90.017365 | 30.892876 |
| N | 31.208178 | 90.605015 | 32.119852 |
| N | 30.658271 | 90.683441 | 29.852121 |
| C | 24.390575 | 88.371184 | 31.887569 |
| C | 25.070647 | 88.113864 | 30.524849 |
| C | 26.264655 | 89.047508 | 30.313823 |
| C | 25.482394 | 86.642460 | 30.376299 |
| S | 28.353163 | 94.323769 | 29.589959 |
| O | 28.176313 | 95.143439 | 30.892893 |
| O | 29.918364 | 94.002309 | 29.465082 |
| O | 28.218313 | 95.407256 | 28.339911 |
| N | 30.251099 | 99.472994 | 19.648377 |
| C | 31.426638 | 100.054480 | 19.714268 |
| N | 32.004270 | 100.542754 | 18.575404 |
| N | 32.131949 | 100.148584 | 20.883965 |
| C | 31.667013 | 99.623555 | 22.111604 |
| O | 32.421183 | 99.731658 | 23.115047 |
| N | 29.806706 | 98.474642 | 23.242945 |
| C | 28.725214 | 97.493467 | 22.962584 |
| C | 27.774132 | 97.944587 | 21.829351 |
| N | 28.528743 | 98.353363 | 20.684157 |
| C | 30.386176 | 99.024620 | 22.054029 |
| C | 29.738725 | 98.970186 | 20.807817 |
| C | 27.875228 | 97.222061 | 24.179599 |
| S | 28.551527 | 96.149596 | 25.423030 |
| C | 26.622171 | 97.717856 | 24.296405 |
| S | 25.601408 | 97.283727 | 25.686504 |
| C | 25.992623 | 98.627822 | 23.252596 |
| O | 26.970550 | 99.040044 | 22.274183 |
| C | 25.383898 | 99.907904 | 23.814719 |
| Mo | 26.697014 | 95.510732 | 26.879130 |
| O | 33.595536 | 92.425970 | 25.739690 |
| O | 31.451619 | 93.476182 | 31.596777 |
| O | 26.088567 | 94.027852 | 26.244045 |
| H | 27.144471 | 102.003672 | 26.944574 |
| H | 28.227475 | 101.896684 | 28.341852 |
| H | 26.559142 | 99.991208 | 28.121877 |
| H | 27.314479 | 99.517829 | 26.605716 |
| H | 28.755477 | 99.550430 | 29.334473 |
| H | 27.949466 | 98.150079 | 28.622063 |
| H | 29.545774 | 98.423215 | 26.593171 |
| H | 30.514483 | 99.485494 | 27.637196 |
| H | 29.731157 | 96.654055 | 28.227895 |
| H | 33.083876 | 98.290129 | 29.797673 |
| H | 32.197618 | 99.352380 | 28.769425 |
| H | 31.026418 | 95.446265 | 29.550317 |
| H | 32.306939 | 96.215689 | 30.438053 |
| H | 30.349044 | 102.445370 | 27.794359 |
| H | 31.217840 | 101.830149 | 25.013412 |
| H | 32.036596 | 103.023627 | 26.057872 |
| H | 32.825780 | 100.324789 | 24.726477 |
| H | 35.137298 | 100.044460 | 26.196554 |
| H | 34.035036 | 98.945649 | 27.037004 |
| H | 34.408003 | 98.823457 | 23.986925 |
| H | 35.578308 | 98.072414 | 25.077811 |
| H | 31.741199 | 97.689969 | 24.349859 |
| H | 35.110958 | 95.504434 | 25.770148 |

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|---|---|---|---|
| H | 30.979899 | 95.305870 | 24.846090 |
| H | 33.107081 | 93.936935 | 25.572376 |
| H | 21.921551 | 95.882448 | 26.797557 |
| H | 23.524798 | 95.361117 | 26.234114 |
| H | 23.586172 | 97.471766 | 27.634310 |
| H | 22.904031 | 96.565098 | 29.007736 |
| H | 24.273598 | 93.301997 | 26.887611 |
| H | 22.225459 | 91.521992 | 27.966274 |
| H | 23.985783 | 89.766615 | 27.723245 |
| H | 23.814246 | 90.723134 | 26.229834 |
| H | 25.198176 | 91.016271 | 27.312723 |
| H | 24.458924 | 93.544849 | 29.558620 |
| H | 24.608160 | 91.899360 | 32.035056 |
| H | 25.012146 | 93.639139 | 31.839877 |
| H | 23.311824 | 93.109037 | 31.956293 |
| H | 23.612056 | 92.134219 | 37.816163 |
| H | 22.734104 | 92.916775 | 36.486974 |
| H | 23.177209 | 91.202203 | 36.366286 |
| H | 24.724713 | 92.629533 | 34.974561 |
| H | 25.164738 | 93.580243 | 36.398243 |
| H | 26.190520 | 91.603735 | 37.478509 |
| H | 25.632227 | 90.546532 | 36.187898 |
| H | 27.602539 | 92.875918 | 35.781879 |
| H | 28.095822 | 91.208244 | 36.101288 |
| H | 26.997829 | 90.546496 | 33.996874 |
| H | 27.567154 | 94.148758 | 34.226176 |
| H | 27.803615 | 94.201236 | 32.464049 |
| H | 27.272056 | 90.816350 | 31.780569 |
| H | 27.335879 | 92.399380 | 31.135227 |
| H | 31.919074 | 92.336280 | 35.512763 |
| H | 30.290584 | 91.965781 | 34.911180 |
| H | 31.416947 | 92.890500 | 33.872318 |
| H | 30.335087 | 94.892169 | 33.690795 |
| H | 30.392616 | 96.698750 | 35.896919 |
| H | 28.691045 | 96.752930 | 33.342584 |
| H | 28.685499 | 98.043835 | 34.536835 |
| H | 31.175657 | 97.105175 | 33.012527 |
| H | 29.488572 | 98.196866 | 31.536523 |
| H | 29.386719 | 99.582890 | 32.639244 |
| H | 30.898071 | 99.267280 | 31.756202 |
| H | 31.829948 | 98.107299 | 35.212727 |
| H | 32.219013 | 99.219993 | 33.894549 |
| H | 30.781810 | 99.512419 | 34.892132 |
| H | 29.125501 | 97.227345 | 37.462024 |
| H | 27.070219 | 95.376403 | 38.270215 |
| H | 27.650507 | 96.806640 | 39.164566 |
| H | 25.847051 | 96.226712 | 36.100992 |
| H | 23.386193 | 97.406804 | 37.053716 |
| H | 24.354577 | 98.656835 | 36.275584 |
| H | 23.454798 | 96.008696 | 34.979142 |
| H | 22.709237 | 97.584783 | 34.742667 |
| H | 25.357189 | 95.313481 | 33.255497 |
| H | 26.666289 | 96.448566 | 31.433349 |
| H | 23.850010 | 100.208598 | 34.163022 |
| H | 27.022385 | 99.082062 | 30.431601 |
| H | 24.760528 | 101.982634 | 32.696470 |
| H | 26.326633 | 101.439435 | 30.847970 |
| H | 32.482191 | 86.571047 | 24.993544 |
| H | 31.945944 | 86.659717 | 26.688190 |
| H | 33.551759 | 88.536262 | 25.800192 |

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|---|---|---|---|
| H | 32.164333 | 89.177552 | 24.933344 |
| H | 29.841368 | 87.926280 | 26.927709 |
| H | 27.749951 | 86.696107 | 25.220097 |
| H | 27.514807 | 87.751550 | 26.627049 |
| H | 28.231599 | 88.541538 | 23.746078 |
| H | 26.736657 | 88.917104 | 24.618921 |
| H | 27.419175 | 90.133023 | 27.102749 |
| H | 29.661036 | 90.389163 | 23.440981 |
| H | 28.578158 | 92.125515 | 27.962301 |
| H | 30.836357 | 92.402646 | 24.305091 |
| H | 30.277819 | 93.640433 | 27.602149 |
| H | 29.692262 | 85.939941 | 34.188610 |
| H | 30.090337 | 87.669409 | 34.225587 |
| H | 28.401775 | 87.148304 | 34.060848 |
| H | 28.869994 | 86.151552 | 31.822162 |
| H | 29.247066 | 87.864545 | 31.801292 |
| H | 31.429633 | 85.771436 | 32.331540 |
| H | 30.927846 | 86.169143 | 30.690158 |
| H | 31.977708 | 88.234924 | 32.696991 |
| H | 32.974663 | 87.418066 | 31.490621 |
| H | 31.561737 | 88.476316 | 29.693902 |
| H | 31.831504 | 90.203973 | 32.812309 |
| H | 31.161072 | 91.634209 | 32.113329 |
| H | 30.784916 | 90.212632 | 28.924622 |
| H | 30.557862 | 91.696277 | 29.883586 |
| H | 23.525881 | 87.706215 | 32.026630 |
| H | 25.094715 | 88.171518 | 32.715987 |
| H | 24.025087 | 89.404458 | 31.948484 |
| H | 24.325976 | 88.348670 | 29.747041 |
| H | 27.028933 | 88.873141 | 31.096300 |
| H | 26.759404 | 88.873350 | 29.346466 |
| H | 25.933409 | 90.097180 | 30.336090 |
| H | 26.214329 | 86.357153 | 31.150627 |
| H | 24.614512 | 85.975113 | 30.481415 |
| H | 25.940043 | 86.444415 | 29.394920 |
| H | 31.389168 | 100.568907 | 17.767179 |
| H | 32.683210 | 101.294448 | 18.635393 |
| H | 33.071734 | 100.542262 | 20.911447 |
| H | 29.444746 | 99.226587 | 23.848319 |
| H | 29.203980 | 96.554829 | 22.633190 |
| H | 27.123437 | 97.096833 | 21.541369 |
| H | 28.057447 | 98.466469 | 19.789623 |
| H | 25.203830 | 98.045680 | 22.729465 |
| H | 24.583046 | 99.673924 | 24.528157 |
| H | 26.158715 | 100.499383 | 24.320998 |
| H | 24.961341 | 100.502402 | 22.993584 |
| H | 33.159208 | 91.678585 | 26.286731 |
| H | 33.999926 | 91.981118 | 24.977638 |
| H | 32.327666 | 93.443037 | 31.174237 |
| H | 30.817387 | 93.716583 | 30.850816 |

The *g*-tensor orientation is given by the following points:

| | | | |
|---|---|---|---|
| origin | 28.430554328 | 94.923602414 | 28.894595035 |
| gx | 28.046829966 | 95.536494046 | 28.567205930 |
| gy | 27.944664637 | 94.954277591 | 29.521519597 |
| gz | 28.927274558 | 95.427077384 | 29.254936724 |

Euler rotation of the hyperfine tensor to the *g*-tensor:

| Atom | Alpha | Beta [degrees] | Gamma | Ax | Ay [MHz] | Az |
|---|---|---|---|---|---|---|
| 121Mo | -21.6 | 70.9 | 42.9 | 143.08 | 47.41 | 51.41 |
| 124O | -4.8 | 76.0 | -6.2 | -0.77 | 3.38 | 8.23 |
| 101O | 26.2 | 9.8 | -5.3 | -2.68 | -28.84 | -1.75 |
| 98S | -15.9 | 6.6 | -60.0 | 1.22 | 3.06 | 1.46 |
| 99O | -19.5 | 69.0 | 31.5 | -0.04 | -0.11 | -4.97 |
| 100O | -25.7 | 30.0 | 43.2 | 0.35 | -0.02 | -0.63 |

Euler rotation of the Electric Field Gradient (EFG) tensor to the *g*-tensor:

| Atom | Alpha | Beta [degrees] | Gamma | EFGx | EFGy [a.u.**-3] | EFGz |
|---|---|---|---|---|---|---|
| 121Mo | -8.6 | 63.2 | 2.7 | 1.29 | -1.05 | -0.24 |
| 124O | -28.9 | 75.5 | -24.0 | -0.18 | 0.00 | 0.18 |
| 101O | 18.2 | 17.6 | -77.4 | -1.07 | 1.56 | -0.49 |
| 98S | 5.2 | 75.0 | 17.9 | -2.16 | 0.84 | 1.32 |
| 99O | -0.3 | 37.1 | -84.9 | 1.56 | -0.56 | -1.00 |
| 100O | 19.0 | 62.3 | -46.8 | -0.92 | -0.70 | 1.62 |

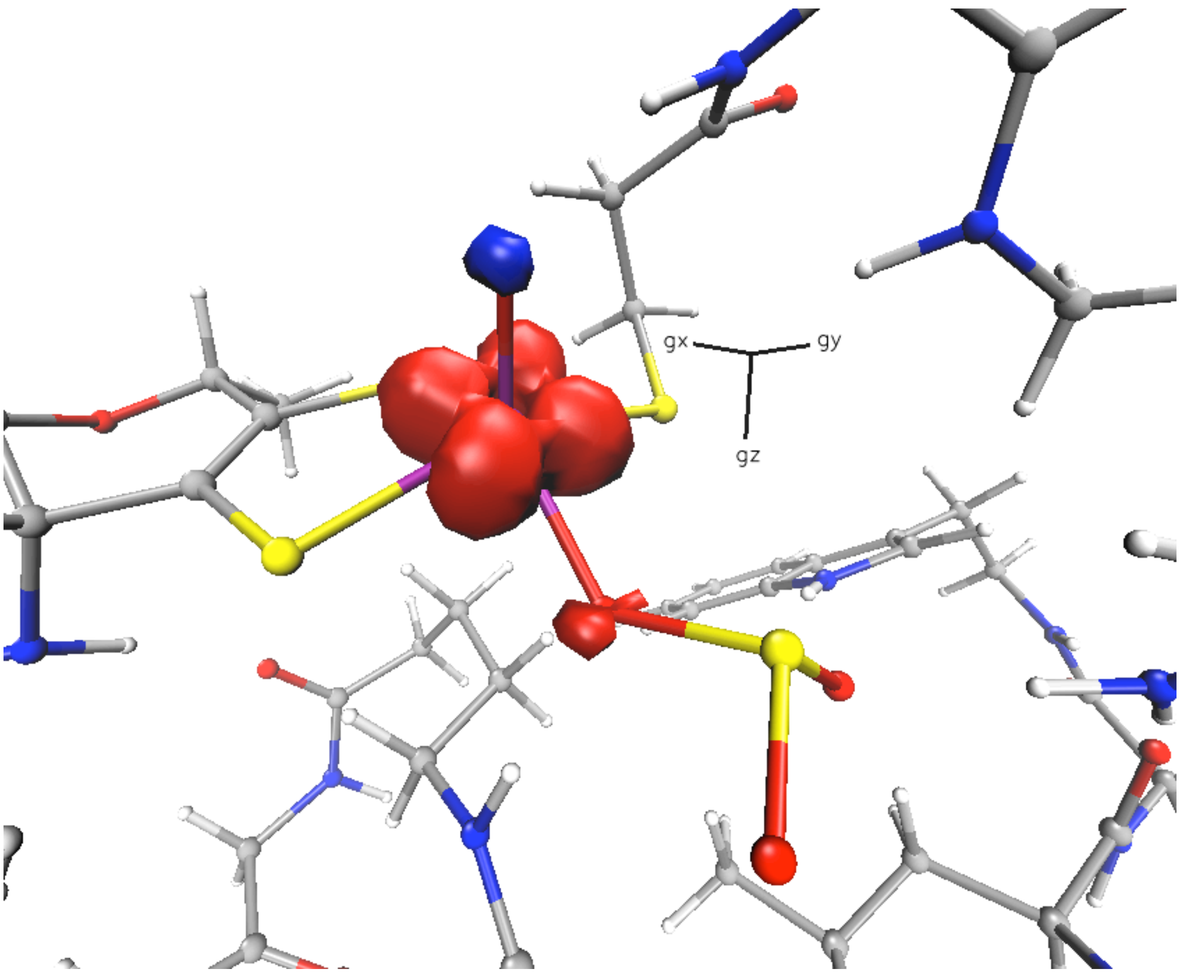


**Figure S8**. The *g*-tensor orientation and total spin density of **2** (contoured at 0.01 e/bohr$^3$).

(3) SO-$SO_3$ (258 atoms):

| | | | |
|---|---|---|---|
| C | 27.975563 | 101.424553 | 27.379790 |
| C | 29.143022 | 101.527538 | 26.393408 |
| O | 29.070837 | 101.029336 | 25.262394 |
| C | 27.502824 | 99.970677 | 27.547640 |
| C | 28.440539 | 99.039067 | 28.332717 |
| C | 29.803119 | 98.806752 | 27.660049 |
| N | 30.430962 | 97.548506 | 28.083671 |
| C | 31.581534 | 97.373504 | 28.740319 |
| N | 32.431943 | 98.384821 | 29.001267 |
| N | 31.903281 | 96.132754 | 29.165937 |
| N | 30.276982 | 102.161107 | 26.826707 |
| C | 31.498592 | 102.062948 | 26.050387 |
| C | 32.441999 | 101.008792 | 26.613346 |
| O | 32.594756 | 100.900078 | 27.851034 |
| N | 33.100046 | 100.266827 | 25.706350 |
| C | 34.234244 | 99.425623 | 26.069707 |
| C | 34.526192 | 98.324108 | 25.025982 |
| C | 33.680888 | 97.091791 | 25.113818 |
| N | 32.349776 | 97.003677 | 24.714583 |
| C | 34.008075 | 95.836435 | 25.575018 |
| C | 31.894647 | 95.753081 | 24.929534 |
| N | 32.890620 | 95.031352 | 25.455642 |
| C | 22.819436 | 95.470347 | 27.131015 |
| C | 22.552306 | 94.123538 | 27.799833 |
| O | 21.627133 | 93.968091 | 28.599927 |
| C | 23.502306 | 96.455562 | 28.084015 |
| S | 25.213929 | 95.939396 | 28.617494 |
| N | 23.435800 | 93.123567 | 27.490987 |
| C | 23.292387 | 91.785722 | 28.054393 |
| C | 23.510406 | 91.711689 | 29.571994 |
| O | 23.042486 | 90.769957 | 30.219208 |
| C | 24.188265 | 90.796368 | 27.306827 |
| N | 24.261650 | 92.698503 | 30.128139 |
| C | 24.309371 | 92.845678 | 31.567957 |
| C | 23.521742 | 92.186296 | 36.722390 |
| C | 24.859746 | 92.575558 | 36.070005 |
| C | 25.985322 | 91.603626 | 36.446161 |
| C | 27.320354 | 91.859005 | 35.737966 |
| N | 27.316418 | 91.481016 | 34.312915 |
| C | 27.391763 | 92.290917 | 33.235262 |
| N | 27.447788 | 93.621236 | 33.341191 |
| N | 27.443163 | 91.748954 | 32.004844 |
| C | 31.091476 | 92.720767 | 34.906002 |
| C | 30.562003 | 93.996645 | 35.556898 |
| O | 30.404893 | 94.097577 | 36.777631 |
| N | 30.223613 | 95.010998 | 34.701245 |
| C | 29.647443 | 96.246228 | 35.222794 |
| C | 28.422341 | 95.942154 | 36.074945 |
| O | 27.514846 | 95.165079 | 35.697621 |
| C | 29.283183 | 97.231413 | 34.098388 |
| C | 30.476454 | 97.819445 | 33.316945 |
| C | 29.944029 | 98.714954 | 32.188657 |
| C | 31.445012 | 98.597704 | 34.218775 |
| N | 28.348422 | 96.635679 | 37.220562 |
| C | 27.287113 | 96.454124 | 38.196159 |
| C | 25.986811 | 97.218242 | 37.880129 |
| O | 25.526560 | 98.041833 | 38.671733 |

| | | | |
|---|---|---|---|
| N | 25.396115 | 96.893234 | 36.698852 |
| C | 24.172027 | 97.570849 | 36.298618 |
| C | 23.606132 | 97.111433 | 34.955304 |
| C | 24.441711 | 97.379688 | 33.736698 |
| C | 24.814936 | 96.408237 | 32.833048 |
| C | 24.897641 | 98.639714 | 33.184995 |
| N | 25.498634 | 96.966497 | 31.768005 |
| C | 25.527922 | 98.337812 | 31.937202 |
| C | 24.819515 | 99.993986 | 33.573035 |
| C | 26.035316 | 99.323945 | 31.085230 |
| C | 25.340232 | 100.978397 | 32.732828 |
| C | 25.938299 | 100.651318 | 31.493718 |
| C | 31.879266 | 87.150592 | 25.706032 |
| C | 30.446645 | 87.097550 | 25.169658 |
| O | 30.186677 | 86.766338 | 24.007328 |
| C | 32.389398 | 88.585582 | 25.947916 |
| C | 31.918585 | 89.242604 | 27.249516 |
| O | 31.324602 | 88.521137 | 28.109174 |
| O | 32.170025 | 90.487206 | 27.424552 |
| N | 29.490304 | 87.463663 | 26.071587 |
| C | 28.091328 | 87.627059 | 25.700067 |
| C | 27.836501 | 88.756145 | 24.677720 |
| C | 28.408035 | 90.082818 | 25.114733 |
| C | 28.017176 | 90.666733 | 26.328187 |
| C | 29.379809 | 90.750135 | 24.354946 |
| C | 28.575869 | 91.858390 | 26.782060 |
| C | 29.945092 | 91.952204 | 24.788658 |
| C | 29.551469 | 92.504025 | 26.011423 |
| O | 30.153313 | 93.670943 | 26.440457 |
| C | 29.434443 | 86.918961 | 33.758664 |
| C | 29.570683 | 86.899603 | 32.226880 |
| C | 31.014093 | 87.004650 | 31.699970 |
| C | 31.655321 | 88.402425 | 31.754220 |
| N | 31.143288 | 89.294819 | 30.706715 |
| C | 30.986989 | 90.623498 | 30.810366 |
| N | 30.895891 | 91.233316 | 32.015262 |
| N | 30.888459 | 91.369118 | 29.702242 |
| C | 24.391353 | 88.371950 | 31.889003 |
| C | 25.071318 | 88.114664 | 30.526223 |
| C | 26.247892 | 89.062933 | 30.296029 |
| C | 25.504501 | 86.648000 | 30.391103 |
| S | 28.422292 | 95.058612 | 29.711888 |
| O | 27.453080 | 94.058229 | 30.359913 |
| O | 29.868711 | 94.377261 | 29.721116 |
| O | 28.187342 | 95.029046 | 28.034191 |
| N | 30.162192 | 99.636203 | 19.642770 |
| C | 31.391595 | 100.093996 | 19.712882 |
| N | 32.004674 | 100.543812 | 18.576891 |
| N | 32.113129 | 100.087530 | 20.879688 |
| C | 31.609236 | 99.564233 | 22.092530 |
| O | 32.375062 | 99.553959 | 23.099070 |
| N | 29.660101 | 98.553411 | 23.195050 |
| C | 28.551921 | 97.623518 | 22.873983 |
| C | 27.597567 | 98.164310 | 21.784075 |
| N | 28.350562 | 98.653216 | 20.667123 |
| C | 30.277402 | 99.098387 | 22.029346 |
| C | 29.613015 | 99.147708 | 20.790759 |
| C | 27.726368 | 97.306868 | 24.094282 |
| S | 28.456977 | 96.229072 | 25.304336 |
| C | 26.472332 | 97.786748 | 24.255481 |

| | | | |
|---|---|---|---|
| S | 25.500684 | 97.312889 | 25.664413 |
| C | 25.820887 | 98.728655 | 23.253059 |
| O | 26.795512 | 99.223345 | 22.309799 |
| C | 25.162036 | 99.955540 | 23.872981 |
| Mo | 26.624748 | 95.459161 | 26.721770 |
| O | 33.291958 | 92.428641 | 25.980229 |
| O | 31.107853 | 94.166658 | 32.014887 |
| O | 25.944106 | 94.057311 | 25.989166 |
| H | 27.152122 | 102.022406 | 26.959348 |
| H | 28.235931 | 101.871704 | 28.353442 |
| H | 26.528047 | 99.982393 | 28.054981 |
| H | 27.321139 | 99.543950 | 26.549986 |
| H | 28.597450 | 99.411566 | 29.358060 |
| H | 27.932596 | 98.066220 | 28.427841 |
| H | 29.671595 | 98.741417 | 26.569865 |
| H | 30.492706 | 99.630893 | 27.870007 |
| H | 29.934983 | 96.703644 | 27.778840 |
| H | 33.113899 | 98.248309 | 29.738566 |
| H | 32.396915 | 99.331972 | 28.573254 |
| H | 31.165248 | 95.392248 | 29.231924 |
| H | 32.880495 | 95.884445 | 29.263335 |
| H | 30.351552 | 102.443695 | 27.800061 |
| H | 31.211782 | 101.841989 | 25.013928 |
| H | 32.035110 | 103.026052 | 26.065219 |
| H | 32.813263 | 100.290248 | 24.712804 |
| H | 35.133007 | 100.059450 | 26.158252 |
| H | 34.057205 | 98.980261 | 27.056984 |
| H | 34.454696 | 98.750665 | 24.013676 |
| H | 35.569830 | 98.007922 | 25.166957 |
| H | 31.808467 | 97.763118 | 24.259369 |
| H | 34.945313 | 95.453450 | 25.961580 |
| H | 30.876494 | 95.408159 | 24.768380 |
| H | 32.868895 | 93.985322 | 25.700920 |
| H | 21.838163 | 95.888371 | 26.859423 |
| H | 23.407344 | 95.364074 | 26.208011 |
| H | 23.588721 | 97.450023 | 27.626464 |
| H | 22.934004 | 96.546507 | 29.019937 |
| H | 24.202222 | 93.307889 | 26.839110 |
| H | 22.240042 | 91.480290 | 27.936892 |
| H | 24.075394 | 89.796370 | 27.743886 |
| H | 23.904189 | 90.746153 | 26.246544 |
| H | 25.244673 | 91.094603 | 27.366619 |
| H | 24.532161 | 93.496589 | 29.549165 |
| H | 24.423966 | 91.860398 | 32.035546 |
| H | 25.160012 | 93.480651 | 31.834406 |
| H | 23.380157 | 93.293816 | 31.959807 |
| H | 23.602610 | 92.175007 | 37.820151 |
| H | 22.723683 | 92.892839 | 36.454312 |
| H | 23.201128 | 91.184629 | 36.399677 |
| H | 24.738082 | 92.592855 | 34.973769 |
| H | 25.146775 | 93.595234 | 36.370813 |
| H | 26.175812 | 91.662325 | 37.530979 |
| H | 25.663524 | 90.564116 | 36.254003 |
| H | 27.601340 | 92.915902 | 35.823254 |
| H | 28.121167 | 91.280851 | 36.223992 |
| H | 27.225117 | 90.488090 | 34.113774 |
| H | 27.488637 | 94.144841 | 34.236278 |
| H | 27.463968 | 94.142736 | 32.460440 |
| H | 27.219629 | 90.771173 | 31.843044 |
| H | 27.450199 | 92.382553 | 31.191506 |

| | | | |
|---|---|---|---|
| H | 31.614711 | 92.139184 | 35.673600 |
| H | 30.236595 | 92.134744 | 34.538076 |
| H | 31.763905 | 92.929900 | 34.066917 |
| H | 30.376182 | 94.884336 | 33.693262 |
| H | 30.394373 | 96.718921 | 35.885843 |
| H | 28.586012 | 96.738444 | 33.401099 |
| H | 28.718582 | 98.063962 | 34.550092 |
| H | 31.031338 | 96.982995 | 32.853029 |
| H | 29.282127 | 98.154889 | 31.511917 |
| H | 29.361948 | 99.556508 | 32.595576 |
| H | 30.769767 | 99.136038 | 31.595180 |
| H | 31.922424 | 97.960145 | 34.977511 |
| H | 32.251430 | 99.053332 | 33.625350 |
| H | 30.921958 | 99.414441 | 34.741736 |
| H | 29.155293 | 97.188824 | 37.493212 |
| H | 27.071282 | 95.377306 | 38.280716 |
| H | 27.638785 | 96.817876 | 39.166652 |
| H | 25.847564 | 96.211965 | 36.083374 |
| H | 23.413574 | 97.414074 | 37.083245 |
| H | 24.360692 | 98.656578 | 36.281411 |
| H | 23.380148 | 96.033488 | 35.001343 |
| H | 22.630249 | 97.615823 | 34.847699 |
| H | 24.613926 | 95.342459 | 32.860950 |
| H | 25.640854 | 96.504129 | 30.865091 |
| H | 24.346444 | 100.279874 | 34.513824 |
| H | 26.466773 | 99.052294 | 30.123653 |
| H | 25.270503 | 102.026031 | 33.031582 |
| H | 26.310414 | 101.449917 | 30.849805 |
| H | 32.503914 | 86.659110 | 24.948857 |
| H | 31.956394 | 86.581548 | 26.644385 |
| H | 33.492460 | 88.580872 | 25.988620 |
| H | 32.116178 | 89.247048 | 25.110434 |
| H | 29.820181 | 87.788648 | 26.983913 |
| H | 27.719671 | 86.681034 | 25.275194 |
| H | 27.532192 | 87.811686 | 26.627966 |
| H | 28.275356 | 88.456651 | 23.716909 |
| H | 26.745730 | 88.832186 | 24.527172 |
| H | 27.262422 | 90.171974 | 26.943345 |
| H | 29.700674 | 90.317740 | 23.404603 |
| H | 28.260867 | 92.292949 | 27.731821 |
| H | 30.700367 | 92.461391 | 24.188121 |
| H | 29.594465 | 94.050771 | 27.161691 |
| H | 29.978362 | 86.078705 | 34.215507 |
| H | 29.834654 | 87.844933 | 34.203807 |
| H | 28.381200 | 86.834761 | 34.062710 |
| H | 29.133637 | 85.962747 | 31.847665 |
| H | 28.966058 | 87.707269 | 31.779953 |
| H | 31.661574 | 86.321397 | 32.274371 |
| H | 31.054547 | 86.654442 | 30.656379 |
| H | 31.487508 | 88.846546 | 32.747809 |
| H | 32.748885 | 88.304055 | 31.639244 |
| H | 31.184710 | 88.945524 | 29.704452 |
| H | 30.876466 | 90.689148 | 32.865970 |
| H | 30.975369 | 92.255169 | 32.081629 |
| H | 31.289737 | 91.022746 | 28.792169 |
| H | 30.512429 | 92.318360 | 29.748310 |
| H | 23.544569 | 87.687099 | 32.039103 |
| H | 25.103807 | 88.201799 | 32.716937 |
| H | 24.001065 | 89.396563 | 31.940364 |
| H | 24.317744 | 88.328764 | 29.750756 |

| | | | |
|---|---|---|---|
| H | 27.022035 | 88.905192 | 31.071895 |
| H | 26.741030 | 88.885693 | 29.328057 |
| H | 25.908413 | 90.109757 | 30.311431 |
| H | 26.248972 | 86.383683 | 31.160938 |
| H | 24.648040 | 85.968714 | 30.511444 |
| H | 25.956589 | 86.444779 | 29.407849 |
| H | 31.389432 | 100.654566 | 17.776152 |
| H | 32.778436 | 101.197063 | 18.636564 |
| H | 33.088208 | 100.382700 | 20.904305 |
| H | 29.324048 | 99.300214 | 23.820627 |
| H | 29.000623 | 96.693508 | 22.482637 |
| H | 26.940456 | 97.343688 | 21.435257 |
| H | 27.889175 | 98.801702 | 19.772902 |
| H | 25.057633 | 98.150014 | 22.689716 |
| H | 24.360476 | 99.658135 | 24.561376 |
| H | 25.908472 | 100.545874 | 24.421123 |
| H | 24.730967 | 100.578080 | 23.077422 |
| H | 32.818255 | 91.696803 | 26.507255 |
| H | 33.693737 | 91.959444 | 25.230579 |
| H | 31.989962 | 94.543887 | 31.862552 |
| H | 30.592928 | 94.320246 | 31.152498 |

The *g*-tensor orientation is given by the following points:

| | | | |
|---|---|---|---|
| origin | 28.364622502 | 94.966271169 | 28.866944020 |
| gx | 28.098462743 | 95.631754834 | 28.525836061 |
| gy | 27.792341695 | 95.018076341 | 29.414552519 |
| gz | 28.845993280 | 95.395819630 | 29.329366400 |

Euler rotation of the hyperfine tensor to the *g*-tensor:

| Atom | Alpha | Beta [degrees] | Gamma | Ax | Ay [MHz] | Az |
|---|---|---|---|---|---|---|
| 121Mo | -15.1 | 62.5 | 27.9 | 143.18 | 47.99 | 51.44 |
| 124O | 20.9 | 74.2 | -23.6 | -0.58 | 1.72 | 8.23 |
| 101O | -15.4 | 39.9 | 15.6 | -9.21 | -25.13 | -8.65 |
| 98S | 12.9 | 29.4 | 73.2 | -4.31 | -4.65 | 3.26 |
| 99O | -15.2 | 41.2 | 47.7 | 1.10 | 1.02 | -5.08 |
| 100O | -23.7 | 69.5 | 28.9 | -1.29 | 0.12 | 0.60 |

Euler rotation of the Electric Field Gradient (EFG) tensor to the *g*-tensor:

| Atom | Alpha | Beta [degrees] | Gamma | EFGx | EFGy [a.u.**-3] | EFGz |
|---|---|---|---|---|---|---|
| 121Mo | -1.9 | 59.1 | -8.2 | 1.28 | -0.87 | -0.42 |
| 124O | 26.1 | 20.0 | 85.2 | 0.14 | 0.01 | -0.15 |
| 101O | -17.7 | 47.3 | 45.5 | -1.05 | -0.51 | 1.55 |
| 98S | -35.2 | 51.9 | 31.9 | -2.19 | 0.82 | 1.38 |
| 99O | 33.2 | 28.9 | 17.2 | 1.51 | -0.99 | -0.52 |
| 100O | 30.2 | 68.8 | -44.3 | -0.94 | -0.68 | 1.62 |

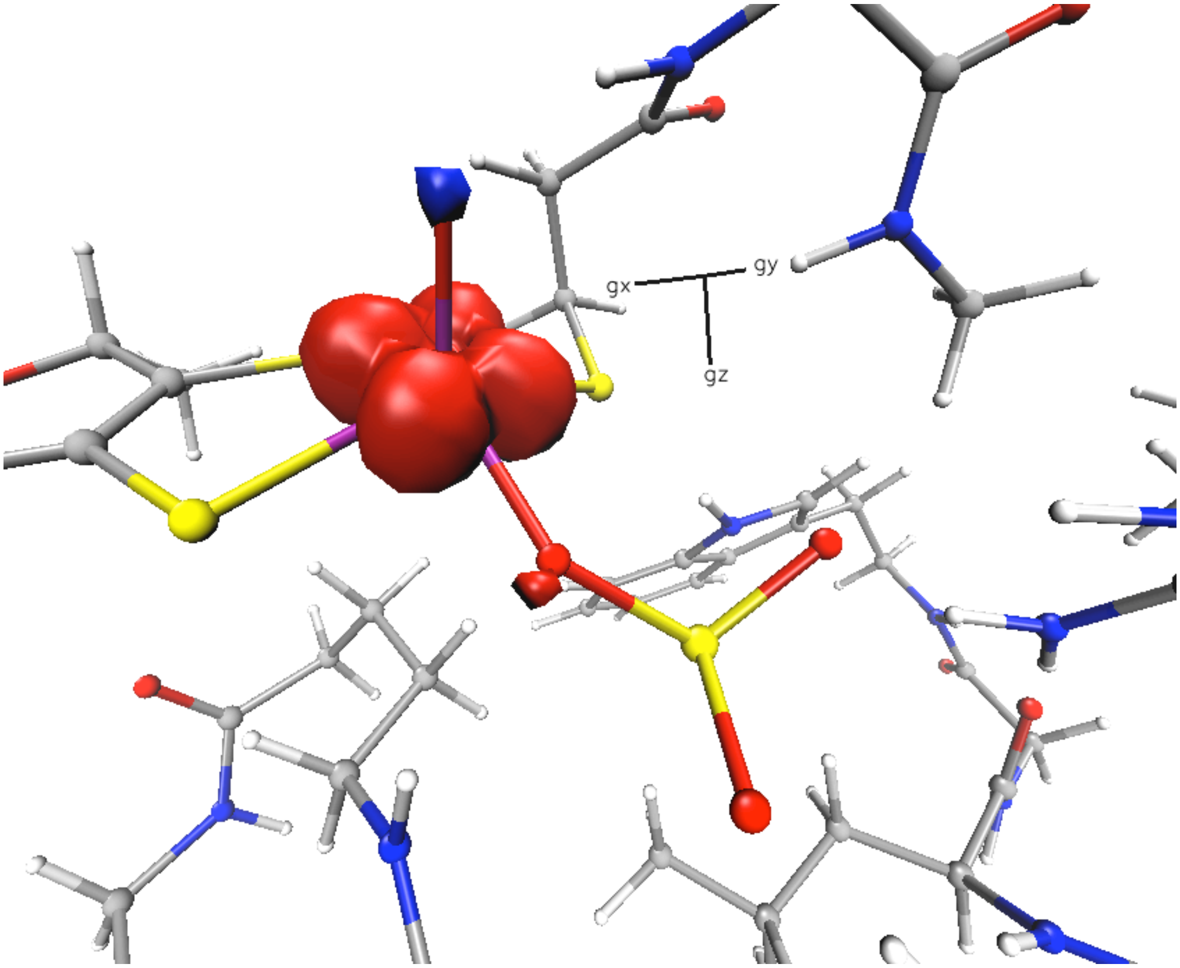


**Figure S9**. The *g*-tensor orientation and total spin density of **3** (contoured at 0.01 e/bohr$^3$).

(4) SO-$SO_3$ (258 atoms):

| | | | |
|---|---|---|---|
| C | 27.977289 | 101.422900 | 27.377645 |
| C | 29.144745 | 101.525978 | 26.391268 |
| O | 29.079945 | 101.000006 | 25.272145 |
| C | 27.559404 | 99.964326 | 27.614395 |
| C | 28.526361 | 99.108544 | 28.445527 |
| C | 29.866147 | 98.770832 | 27.772134 |
| N | 30.497780 | 97.611850 | 28.420549 |
| C | 31.526020 | 97.614403 | 29.271054 |
| N | 32.373219 | 98.649078 | 29.391464 |
| N | 31.743808 | 96.497342 | 30.009962 |
| N | 30.274926 | 102.170975 | 26.817703 |
| C | 31.500209 | 102.061827 | 26.048207 |
| C | 32.450444 | 101.019914 | 26.631654 |
| O | 32.604480 | 100.926728 | 27.872024 |
| N | 33.106025 | 100.278789 | 25.729332 |
| C | 34.236411 | 99.425077 | 26.067979 |
| C | 34.573473 | 98.411891 | 24.947863 |
| C | 33.859079 | 97.091124 | 24.940242 |
| N | 32.566545 | 96.840100 | 24.475110 |
| C | 34.369849 | 95.856585 | 25.278894 |
| C | 32.326404 | 95.514647 | 24.532196 |
| N | 33.407562 | 94.901054 | 25.021170 |

| | | | |
|---|---|---|---|
| C | 23.123507 | 95.450875 | 26.962541 |
| C | 22.720240 | 94.152583 | 27.657416 |
| O | 21.706230 | 94.075567 | 28.353792 |
| C | 23.694432 | 96.488945 | 27.932059 |
| S | 25.340022 | 96.054098 | 28.700563 |
| N | 23.571311 | 93.093450 | 27.471690 |
| C | 23.296115 | 91.783194 | 28.053642 |
| C | 23.506584 | 91.707936 | 29.571900 |
| O | 23.048321 | 90.759020 | 30.216252 |
| C | 24.098987 | 90.699081 | 27.328743 |
| N | 24.237792 | 92.707953 | 30.128746 |
| C | 24.312816 | 92.843900 | 31.567062 |
| C | 23.525234 | 92.185143 | 36.721582 |
| C | 24.863169 | 92.574585 | 36.069161 |
| C | 25.962091 | 91.552078 | 36.385553 |
| C | 27.299113 | 91.805544 | 35.682796 |
| N | 27.293976 | 91.460455 | 34.249155 |
| C | 27.482871 | 92.286791 | 33.197246 |
| N | 27.610796 | 93.607580 | 33.338411 |
| N | 27.566799 | 91.762116 | 31.960643 |
| C | 31.094888 | 92.720917 | 34.905245 |
| C | 30.610867 | 94.010921 | 35.552114 |
| O | 30.517381 | 94.147383 | 36.773542 |
| N | 30.247861 | 95.022493 | 34.694578 |
| C | 29.650113 | 96.246124 | 35.221472 |
| C | 28.432534 | 95.923413 | 36.081345 |
| O | 27.546422 | 95.115052 | 35.723984 |
| C | 29.286620 | 97.246247 | 34.111475 |
| C | 30.481602 | 97.989830 | 33.479279 |
| C | 29.966885 | 98.878501 | 32.338623 |
| C | 31.262590 | 98.829453 | 34.501353 |
| N | 28.348389 | 96.634247 | 37.216047 |
| C | 27.289688 | 96.453982 | 38.194764 |
| C | 25.986749 | 97.213391 | 37.879874 |
| O | 25.526791 | 98.036338 | 38.672300 |
| N | 25.394269 | 96.885803 | 36.700540 |
| C | 24.174402 | 97.569767 | 36.296997 |
| C | 23.614330 | 97.112686 | 34.949407 |
| C | 24.502464 | 97.382727 | 33.773723 |
| C | 25.135021 | 96.405861 | 33.033406 |
| C | 24.869853 | 98.645338 | 33.171281 |
| N | 25.871618 | 96.969646 | 32.015189 |
| C | 25.722123 | 98.337201 | 32.063495 |
| C | 24.568707 | 100.001543 | 33.417255 |
| C | 26.250335 | 99.320926 | 31.220520 |
| C | 25.104170 | 100.983535 | 32.584065 |
| C | 25.940115 | 100.649869 | 31.491656 |
| C | 31.884003 | 87.149498 | 25.706142 |
| C | 30.428943 | 86.931378 | 25.287273 |
| O | 30.123739 | 86.174662 | 24.357251 |
| C | 32.401328 | 88.600507 | 25.621647 |
| C | 32.063588 | 89.534736 | 26.792078 |
| O | 31.307440 | 89.076633 | 27.728303 |
| O | 32.524666 | 90.709798 | 26.802493 |
| N | 29.515175 | 87.630123 | 26.014370 |
| C | 28.095966 | 87.625174 | 25.700037 |
| C | 27.759688 | 88.189983 | 24.295383 |
| C | 28.263734 | 89.596207 | 24.072775 |
| C | 27.628488 | 90.687351 | 24.683550 |
| C | 29.400684 | 89.861573 | 23.291558 |

| | | | |
|---|---|---|---|
| C | 28.111028 | 91.987203 | 24.544320 |
| C | 29.904011 | 91.159790 | 23.144052 |
| C | 29.267888 | 92.222838 | 23.792275 |
| O | 29.789947 | 93.500923 | 23.664138 |
| C | 29.439087 | 86.918589 | 33.758766 |
| C | 29.575359 | 86.899025 | 32.226987 |
| C | 30.983027 | 86.583327 | 31.691100 |
| C | 31.925975 | 87.792557 | 31.569302 |
| N | 31.470410 | 88.762708 | 30.572439 |
| C | 31.015185 | 90.012232 | 30.778931 |
| N | 30.886170 | 90.527947 | 32.034453 |
| N | 30.662334 | 90.738399 | 29.720725 |
| C | 24.395727 | 88.370239 | 31.888793 |
| C | 25.075770 | 88.112886 | 30.526065 |
| C | 26.257017 | 89.058077 | 30.299659 |
| C | 25.503923 | 86.645263 | 30.387007 |
| S | 28.800965 | 94.885732 | 30.042129 |
| O | 29.211856 | 93.369374 | 30.039244 |
| O | 27.475563 | 94.970421 | 30.828436 |
| O | 28.484275 | 95.310289 | 28.468531 |
| N | 30.466366 | 99.175054 | 19.639093 |
| C | 31.511014 | 99.968456 | 19.706980 |
| N | 32.006739 | 100.541653 | 18.574952 |
| N | 32.165244 | 100.220083 | 20.880805 |
| C | 31.784907 | 99.649767 | 22.111309 |
| O | 32.490650 | 99.923844 | 23.115087 |
| N | 30.145582 | 98.225561 | 23.265742 |
| C | 29.120173 | 97.160430 | 23.043528 |
| C | 28.176468 | 97.480620 | 21.860653 |
| N | 28.937152 | 97.809102 | 20.693540 |
| C | 30.636571 | 98.823881 | 22.054624 |
| C | 30.029378 | 98.623035 | 20.805517 |
| C | 28.250668 | 96.952582 | 24.262190 |
| S | 28.891133 | 95.964362 | 25.604565 |
| C | 26.994295 | 97.449653 | 24.322014 |
| S | 25.936918 | 97.115969 | 25.708139 |
| C | 26.375989 | 98.282339 | 23.209169 |
| O | 27.351443 | 98.599664 | 22.195472 |
| C | 25.788840 | 99.613157 | 23.669160 |
| Mo | 26.954844 | 95.404127 | 27.056653 |
| O | 33.780595 | 92.318180 | 25.228566 |
| O | 31.436901 | 93.402903 | 31.547657 |
| O | 26.363323 | 93.893445 | 26.468058 |
| H | 27.133282 | 101.967871 | 26.927604 |
| H | 28.214182 | 101.924332 | 28.330944 |
| H | 26.588216 | 99.967287 | 28.130725 |
| H | 27.385793 | 99.481752 | 26.641353 |
| H | 28.727308 | 99.586679 | 29.418714 |
| H | 28.013093 | 98.157903 | 28.662138 |
| H | 29.706911 | 98.498308 | 26.719585 |
| H | 30.557211 | 99.619723 | 27.803617 |
| H | 29.973774 | 96.728552 | 28.316054 |
| H | 32.989306 | 98.680169 | 30.195575 |
| H | 32.400801 | 99.479753 | 28.760852 |
| H | 30.945445 | 95.892927 | 30.233158 |
| H | 32.563606 | 96.426774 | 30.600506 |
| H | 30.343882 | 102.470202 | 27.786406 |
| H | 31.220533 | 101.819197 | 25.015054 |
| H | 32.032301 | 103.027503 | 26.047314 |
| H | 32.817402 | 100.316055 | 24.734855 |

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|---|---|---|---|
| H | 35.123201 | 100.059556 | 26.233771 |
| H | 34.038046 | 98.903284 | 27.015172 |
| H | 34.446760 | 98.902928 | 23.970603 |
| H | 35.642927 | 98.174561 | 25.042047 |
| H | 31.847224 | 97.519683 | 24.123661 |
| H | 35.348300 | 95.590211 | 25.661566 |
| H | 31.415333 | 95.006343 | 24.228256 |
| H | 33.514561 | 93.815405 | 25.134304 |
| H | 22.200004 | 95.869695 | 26.532840 |
| H | 23.825313 | 95.272999 | 26.135608 |
| H | 23.822789 | 97.458727 | 27.432392 |
| H | 23.022237 | 96.627913 | 28.790235 |
| H | 24.435972 | 93.228629 | 26.942291 |
| H | 22.219847 | 91.586094 | 27.927775 |
| H | 23.877445 | 89.720800 | 27.773011 |
| H | 23.826448 | 90.668683 | 26.264840 |
| H | 25.181055 | 90.885673 | 27.410800 |
| H | 24.483038 | 93.514531 | 29.552080 |
| H | 24.419269 | 91.851996 | 32.022248 |
| H | 25.180255 | 93.463333 | 31.820402 |
| H | 23.401593 | 93.309191 | 31.979470 |
| H | 23.622822 | 92.114309 | 37.815707 |
| H | 22.740991 | 92.924033 | 36.504288 |
| H | 23.174724 | 91.209950 | 36.352383 |
| H | 24.729647 | 92.647308 | 34.976662 |
| H | 25.181836 | 93.569559 | 36.416598 |
| H | 26.158740 | 91.550702 | 37.470789 |
| H | 25.608733 | 90.534297 | 36.142446 |
| H | 27.590034 | 92.856825 | 35.796215 |
| H | 28.092193 | 91.205960 | 36.156965 |
| H | 27.272857 | 90.467908 | 34.030129 |
| H | 27.588815 | 94.097581 | 34.251952 |
| H | 27.570073 | 94.179285 | 32.476197 |
| H | 27.271884 | 90.810889 | 31.760459 |
| H | 27.850143 | 92.353529 | 31.169302 |
| H | 31.889340 | 92.297781 | 35.532095 |
| H | 30.260218 | 92.004105 | 34.883203 |
| H | 31.459408 | 92.881871 | 33.882362 |
| H | 30.263914 | 94.860111 | 33.690283 |
| H | 30.395793 | 96.710042 | 35.890015 |
| H | 28.704553 | 96.728358 | 33.330863 |
| H | 28.602171 | 97.997119 | 34.540025 |
| H | 31.172482 | 97.237925 | 33.052674 |
| H | 29.449422 | 98.286965 | 31.569277 |
| H | 29.245910 | 99.620629 | 32.715134 |
| H | 30.789820 | 99.428616 | 31.858071 |
| H | 31.734524 | 98.217709 | 35.283975 |
| H | 32.067234 | 99.394799 | 34.008417 |
| H | 30.601146 | 99.561750 | 34.992277 |
| H | 29.137040 | 97.220880 | 37.471575 |
| H | 27.077534 | 95.376783 | 38.281784 |
| H | 27.642765 | 96.820788 | 39.163669 |
| H | 25.846763 | 96.211794 | 36.078718 |
| H | 23.415314 | 97.418735 | 37.082289 |
| H | 24.370576 | 98.654331 | 36.273773 |
| H | 23.384879 | 96.035221 | 34.991554 |
| H | 22.645062 | 97.623021 | 34.822145 |
| H | 25.090591 | 95.327121 | 33.151704 |
| H | 26.397758 | 96.429990 | 31.321014 |
| H | 23.917792 | 100.288056 | 34.245697 |

| | | | |
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| H | 26.875347 | 99.041428 | 30.375825 |
| H | 24.866425 | 102.031916 | 32.773081 |
| H | 26.328866 | 101.444130 | 30.852084 |
| H | 32.478055 | 86.511218 | 25.039505 |
| H | 32.024941 | 86.776734 | 26.733578 |
| H | 33.500378 | 88.594511 | 25.535133 |
| H | 32.030837 | 89.089933 | 24.705339 |
| H | 29.882517 | 88.230115 | 26.759965 |
| H | 27.709714 | 86.593595 | 25.756270 |
| H | 27.589750 | 88.216416 | 26.476423 |
| H | 28.186910 | 87.508449 | 23.548144 |
| H | 26.663075 | 88.156211 | 24.182259 |
| H | 26.723054 | 90.523692 | 25.273350 |
| H | 29.900861 | 89.032848 | 22.785373 |
| H | 27.590410 | 92.813659 | 25.030820 |
| H | 30.788569 | 91.352446 | 22.534059 |
| H | 29.290278 | 94.093063 | 24.279137 |
| H | 29.702131 | 85.941883 | 34.191902 |
| H | 30.092627 | 87.670980 | 34.230387 |
| H | 28.405665 | 87.143095 | 34.059698 |
| H | 28.878288 | 86.144941 | 31.830621 |
| H | 29.232726 | 87.857031 | 31.801090 |
| H | 31.474813 | 85.832115 | 32.331770 |
| H | 30.902604 | 86.123634 | 30.693117 |
| H | 32.047027 | 88.289283 | 32.543252 |
| H | 32.931130 | 87.451472 | 31.277141 |
| H | 31.507515 | 88.498342 | 29.574927 |
| H | 31.327052 | 90.022520 | 32.794960 |
| H | 30.964609 | 91.553186 | 32.091272 |
| H | 30.849426 | 90.300589 | 28.777427 |
| H | 30.178423 | 91.633859 | 29.805482 |
| H | 23.552032 | 87.681834 | 32.040540 |
| H | 25.108909 | 88.205613 | 32.717140 |
| H | 24.000674 | 89.393016 | 31.937854 |
| H | 24.324331 | 88.332284 | 29.750499 |
| H | 27.035490 | 88.886414 | 31.068541 |
| H | 26.738408 | 88.892741 | 29.323714 |
| H | 25.922041 | 90.106556 | 30.330308 |
| H | 26.242048 | 86.374953 | 31.161044 |
| H | 24.644236 | 85.968773 | 30.499561 |
| H | 25.961063 | 86.444640 | 29.405525 |
| H | 31.427979 | 100.432480 | 17.747375 |
| H | 32.541058 | 101.402100 | 18.630837 |
| H | 33.013829 | 100.784131 | 20.911998 |
| H | 29.741288 | 98.955677 | 23.869944 |
| H | 29.648053 | 96.224730 | 22.792016 |
| H | 27.553132 | 96.594766 | 21.641736 |
| H | 28.459212 | 97.841450 | 19.795025 |
| H | 25.576458 | 97.669916 | 22.740912 |
| H | 24.977696 | 99.446831 | 24.389983 |
| H | 26.571345 | 100.227262 | 24.136050 |
| H | 25.382577 | 100.149432 | 22.800810 |
| H | 33.276665 | 91.638198 | 25.821284 |
| H | 33.957180 | 91.861042 | 24.390694 |
| H | 32.189561 | 93.233737 | 30.953895 |
| H | 30.632043 | 93.428171 | 30.940886 |

The *g*-tensor orientation is given by the following points:

| | | | |
|---|---|---|---|
| origin | 28.473199361 | 94.874992206 | 28.799572068 |
| gx | 28.927162876 | 94.264131408 | 29.025028000 |
| gy | 28.936746438 | 94.985164533 | 28.164709618 |
| gz | 28.930479800 | 95.369739928 | 29.219314200 |

Euler rotation of the hyperfine tensor to the *g*-tensor:

| Atom | Alpha | Beta [degrees] | Gamma | Ax | Ay [MHz] | Az |
|---|---|---|---|---|---|---|
| 121Mo | -23.7 | 32.7 | 16.6 | 53.55 | 50.07 | 144.83 |
| 124O | -24.4 | 18.5 | 25.8 | 7.81 | 1.81 | -0.12 |
| 101O | -11.2 | 78.5 | 33.1 | -9.49 | -30.16 | -10.37 |
| 98S | 17.0 | 46.8 | -1.2 | 3.44 | -1.35 | -1.84 |
| 99O | -22.0 | 81.7 | 2.0 | -1.49 | 0.37 | -0.21 |
| 100O | -29.2 | 59.8 | -21.2 | 1.16 | -4.93 | 0.40 |

Euler rotation of the Electric Field Gradient (EFG) tensor to the *g*-tensor:

| Atom | Alpha | Beta [degrees] | Gamma | EFGx | EFGy [a.u.**-3] | EFGz |
|---|---|---|---|---|---|---|
| 121Mo | 10.0 | 29.3 | 10.5 | -0.40 | -0.96 | 1.36 |
| 124O | -45.0 | 21.2 | -71.8 | -0.02 | 0.14 | -0.12 |
| 101O | -39.9 | 51.1 | -17.5 | -0.69 | 1.61 | -0.93 |
| 98S | -17.6 | 76.8 | -35.4 | -2.14 | 1.39 | 0.75 |
| 99O | 31.3 | 68.4 | 1.5 | -0.67 | -0.89 | 1.56 |
| 100O | 18.5 | 24.6 | 2.9 | -0.61 | 1.56 | -0.96 |

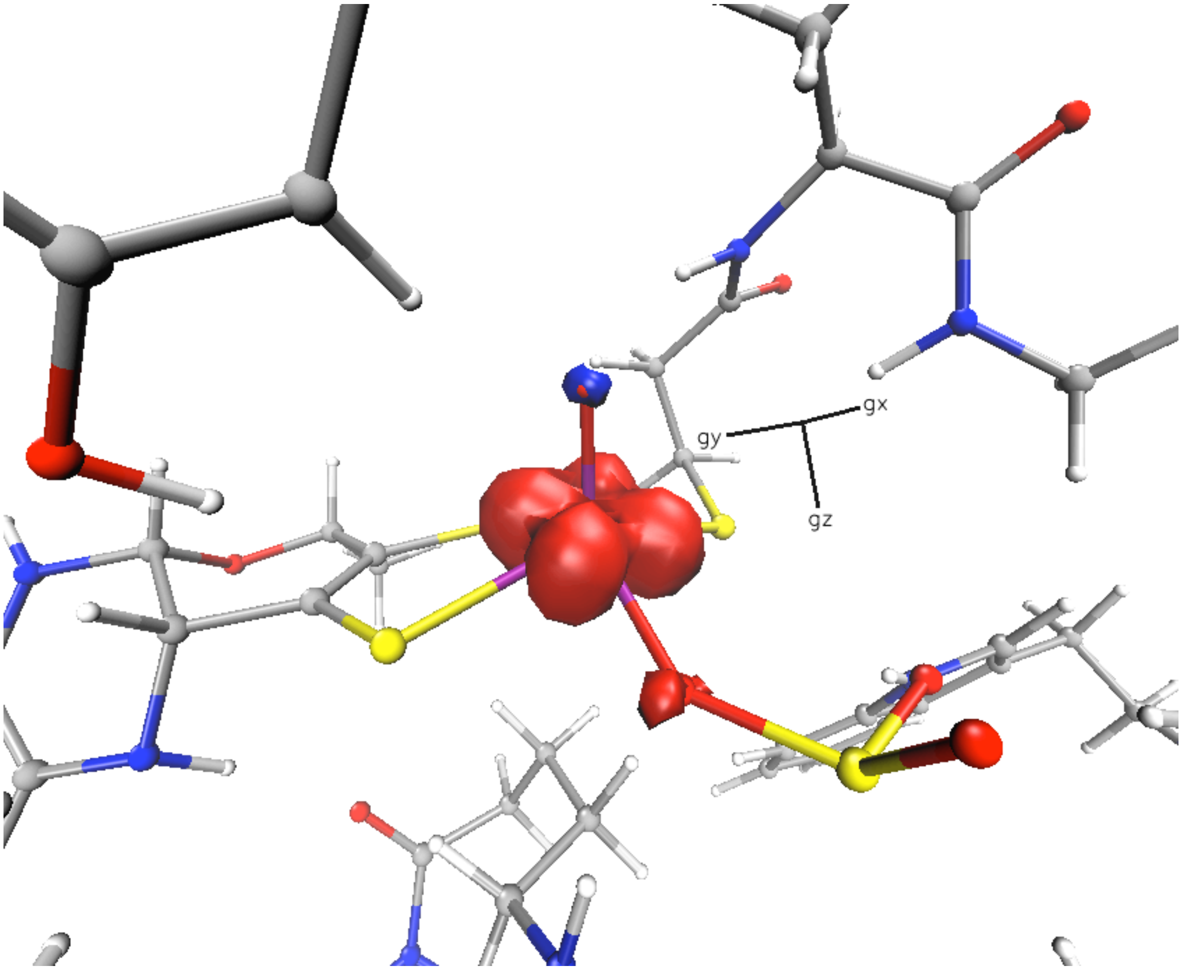


**Figure S10**. The *g*-tensor orientation and total spin density of **4** (contoured at 0.01 e/bohr$^3$).

**Table S1**. Experimental magnetic resonance parameters for *wt* and mutant *blocked* forms of SO and DFT-calculated magnetic resonance parameters for sulfate- and sulfite-bound SO models.

| | | ***hfi*** | | ***nqi*** | | **principal *g*-values** | | | |
|---|---|---|---|---|---|---|---|---|---|
| **Species** | | $a_{iso}$/MHz | ($T_{11}$, $T_{22}$, $T_{33}$)/MHz | $e^2Qq/h$/MHz | $\eta$ | $g_x$ | $g_y$ | $g_z$ | ref |
| | $^{33}$S | 2.1 | (1.6, 2.5, –4.1) | 36 | 0.2 | | | | |
| *blocked* R160Q hSO | $^{17}O_{coordinated}$ | –9 | (5, 5, –10) | 7 | – | 1.951 | 1.971 | 2.006 | this work |
| | $^{17}O_{remote}$ | –4.5 | (1.25, 1.25, –2.5) | 5 | – | | | | |
| *blocked wt* hSO*† | $^{33}$S | 2.6 | (–0.3, –0.3, 0.6) | 36 | – | 1.963 | 1.972 | 1.999 | 10 |
| *blocked* At-SO* | $^{33}$S | 3.3 | (1.3, 1.5, –2.8) | 40 | 0 | 1.963 | 1.974 | 2.005 | 11 |
| *wt lpH* cSO | $^{95,97}$Mo | 98.4‡ | (–48.3, –23.4, 71.7)‡ | – | – | 1.968 | 1.974 | 2.007 | 12 |
| *wt hpH* cSO | $^{95,97}$Mo | 86.7‡ | (–52.8, –23.7, 76.5)‡ | – | – | 1.954 | 1.966 | 1.990 | 12 |
| | $^{95}$Mo | 80.6 | (–32.6, –29.6, 62.2) | –36 | 1.0 | | | | |
| | $^{33}$S | 3.33 | (–0.53, –0.40, 0.92) | 4.4 | 0.7 | | | | |
| | $^{17}O_{coordinated}$ | –9.56 | (6.06, 5.17, –11.22) | 10.9 | 0.3 | | | | |
| (1) SO-Sulfate (DFT) | $^{17}O_{remote}$ | –1.23 (a) | (1.18, 1.17, –2.35) | 10.6 | 0.1 | 1.992 | 1.997 | 2.004 | this work |
| | | 0.02 (b) | (0.00, –0.36, 0.36) | 10.5 | 0.1 | | | | |
| | | 0.01 (c) | (0.00, –0.27, 0.27) | 10.9 | 0.1 | | | | |
| | $^{17}O_{oxo}$ | 3.26 | (–3.71, –2.34, 6.04) | 1.2 | 1.0 | | | | |
| | $^{95}$Mo | 80.6 | (–33.2, –29.2, 62.4) | –36 | 0.6 | | | | |
| | $^{33}$S | 1.91 | (–0.69, –0.45, 1.14) | 27.9 | 0.2 | | | | |
| | $^{17}O_{coordinated}$ | –11.09 | (9.34, 8.41, –17.75) | 9.6 | 0.4 | | | | |
| (2) SO-Sulfite (DFT) | $^{17}O_{remote}$ | –1.71 (a) | (1.66, 1.60, –3.26) | 9.5 | 0.3 | 1.994 | 1.996 | 2.004 | this work |
| | | –0.10 (b) | (0.08, 0.45, –0.53) | 9.9 | 0.1 | | | | |
| | $^{17}O_{oxo}$ | 3.61 | (–4.38, –0.24, 4.62) | 1.1 | 1.0 | | | | |
| | $^{95}$Mo | 80.9 | (–32.9, –29.4, 62.3) | 36 | 0.4 | | | | |
| | $^{33}$S | –1.90 | (5.16, –2.41, –2.75) | 28.4 | 0.3 | | | | |
| | $^{17}O_{coordinated}$ | –14.33 | (5.68, 5.12, –10.80) | 9.5 | 0.3 | | | | |
| (3) SO-Sulfite (DFT) | $^{17}O_{remote}$ | –0.99 (a) | (2.01, 2.09, –4.10) | 9.2 | 0.3 | 1.993 | 1.997 | 2.003 | this work |
| | | –0.19 (b) | (0.30, 0.79, –1.10) | 9.9 | 0.2 | | | | |
| | $^{17}O_{oxo}$ | 3.13 | (–3.70, –1.40, 5.10) | 0.9 | 0.9 | | | | |
| | $^{95}$Mo | 82.8 | (–32.8, –29.3, 62.0) | –38 | 0.4 | | | | |
| | $^{33}$S | 0.09 | (–1.43, –1.93, 3.36) | 27.7 | 0.3 | | | | |
| | $^{17}O_{coordinated}$ | –16.67 | (7.18, 6.30, –13.49) | 9.9 | 0.1 | | | | |
| (4) SO-Sulfite (DFT) | $^{17}O_{remote}$ | –1.13 (a) | (1.53, 2.28, –3.81) | 9.6 | 0.2 | 1.962 | 1.967 | 2.003 | this work |
| | | –0.44 (b) | (0.23, 0.44, –1.04) | 9.5 | 0.1 | | | | |
| | $^{17}O_{oxo}$ | 3.16 | (–3.29, –1.35, 4.64) | 0.9 | 0.7 | | | | |

* The $^{17}$O magnetic resonance parameters for these species have not been fully evaluated.

† The *blocked* form of this enzyme was obtained only under low-pH conditions in the strict absence of chloride.

‡ To our knowledge, these are the only reported Mo hyperfine values for SO. Although the Mo centers of the *lpH* and *hpH* forms differ structurally from the *blocked* form, these are provided here to facilitate general comparisons with the calculated $^{95}$Mo(V) hyperfine values.

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