# Supporting Information

.

## List of Figures:

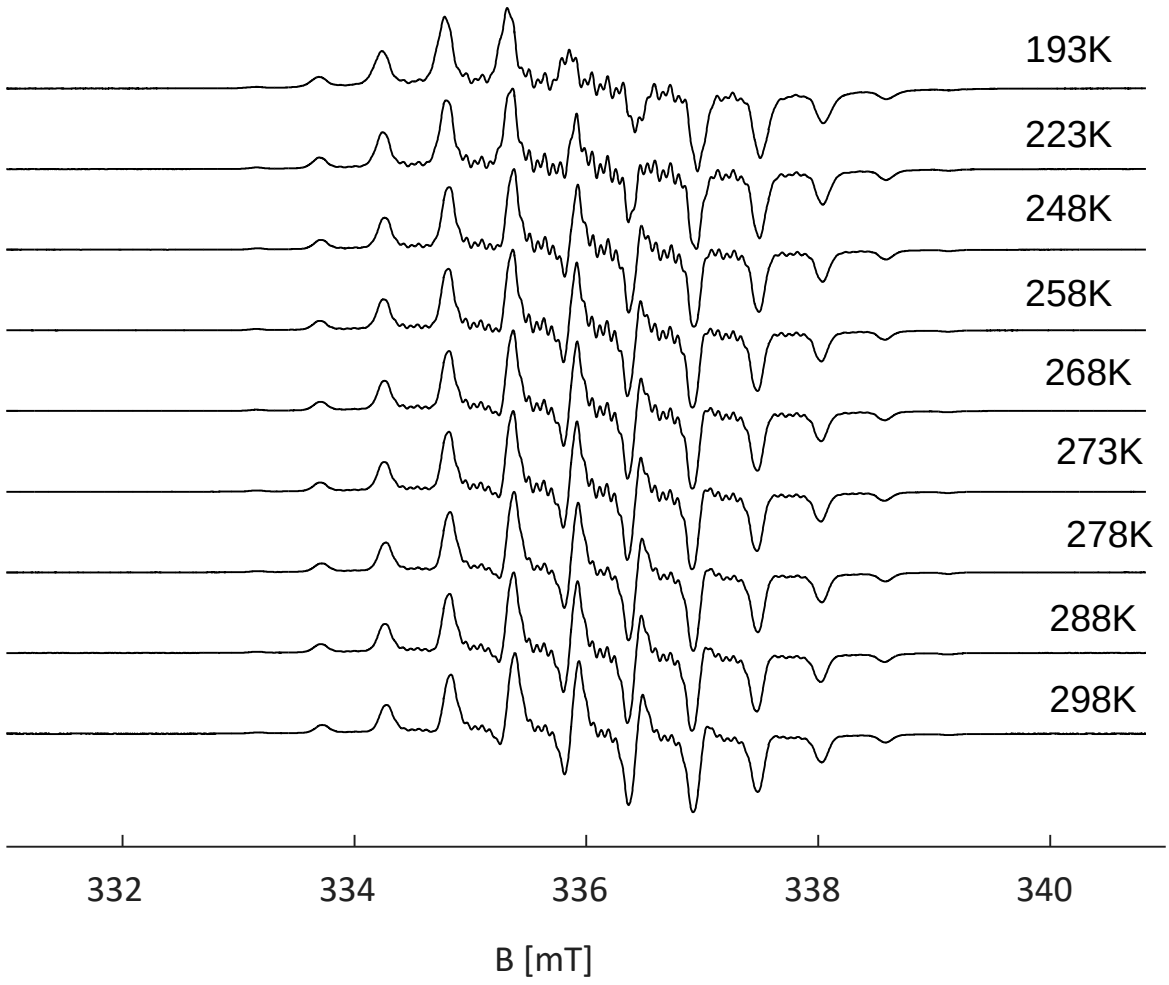



**Figure S1.** X-band CW EPR of Na-DAD complex in THF, recorded at different temperatures.

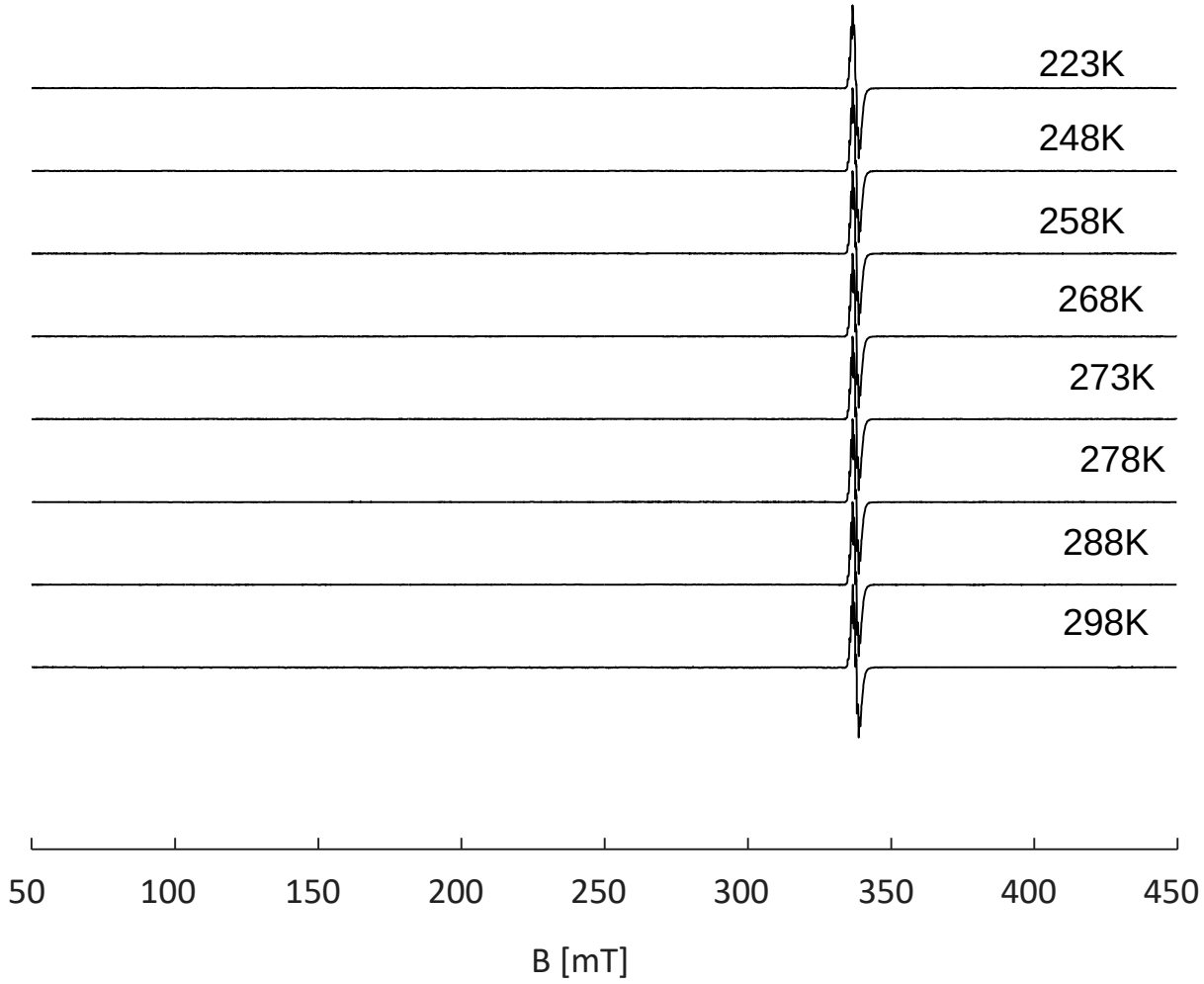



**Figure S2.** The same as FigureS1, but with a long range scan. There is no transition at half field position (~170mT), discarding a singlet to triplet electronic transition.

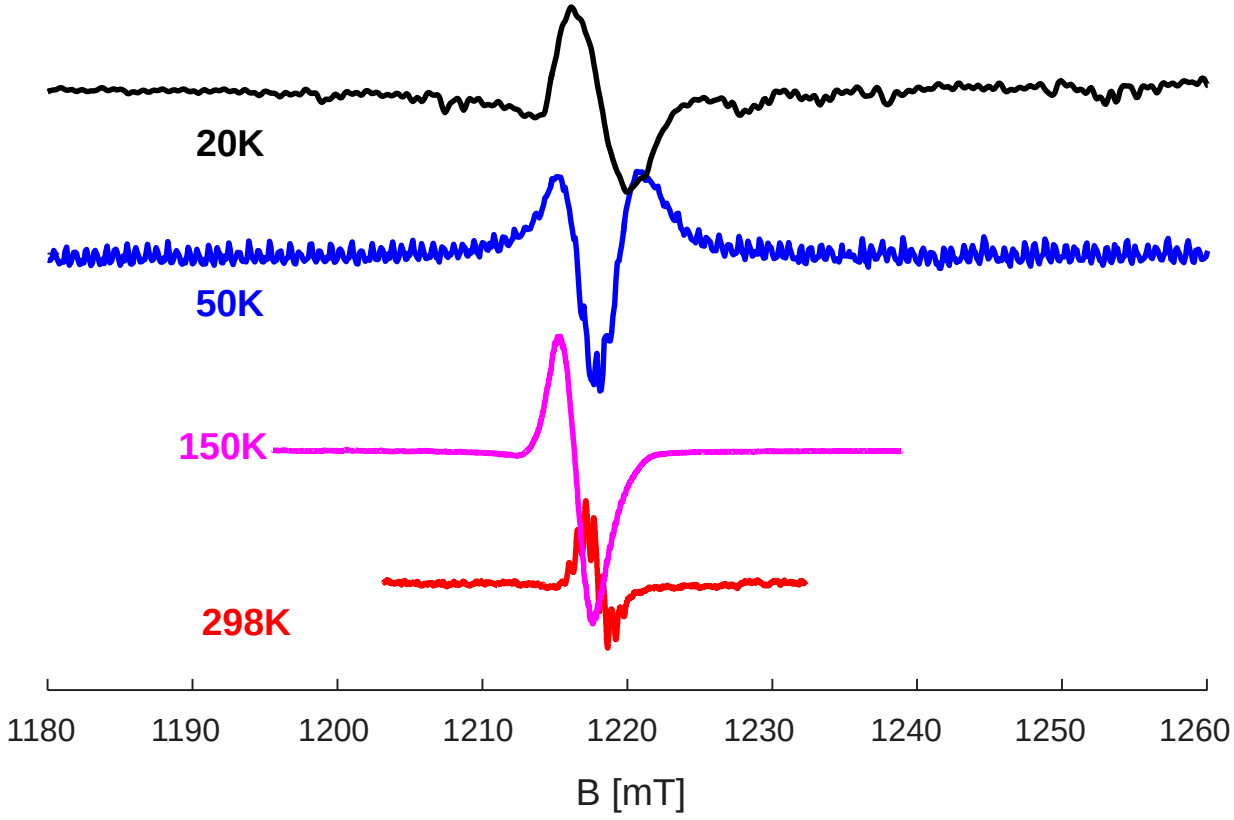
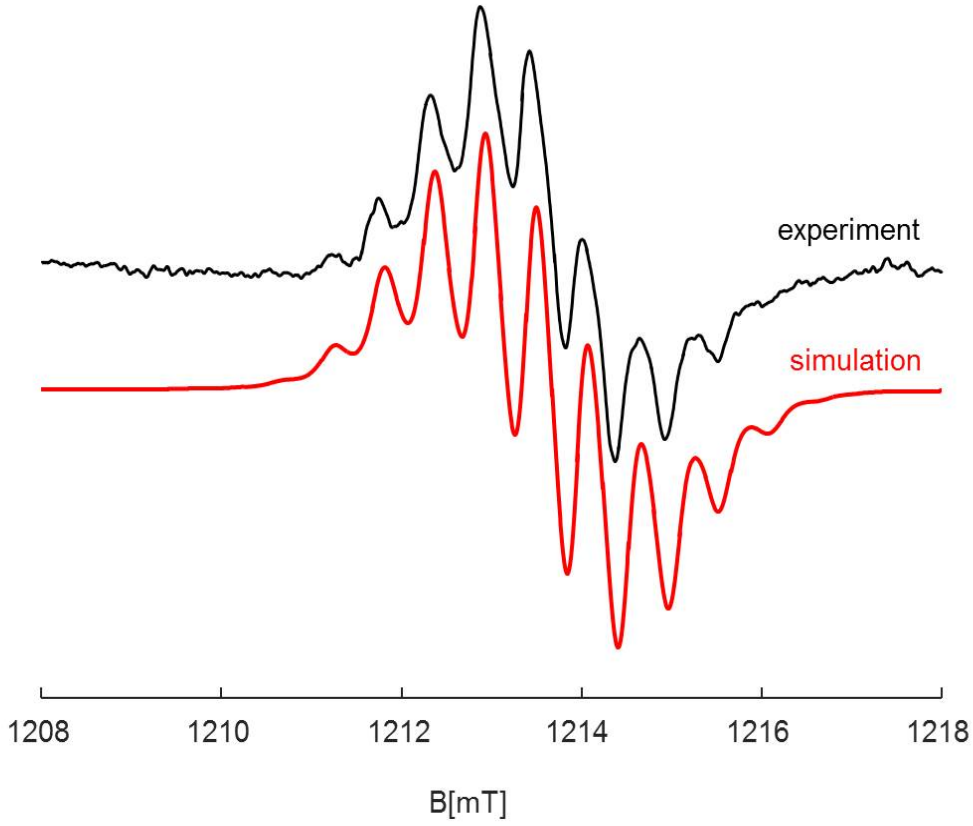



**Figure S3.** CW EPR temperature series of NaDAD complex (dissolved in THF) recorded at 34GHz at 1mW of microwave power and 0.7mT of modulation amplitude.




**FigureS4.**Q-band CW room temperature of Na-DAD THF ( in black) is simulated (in red) at 33.9GHz with $a_{iso}(^{14}N)$=0.53mT(#2) , $a_{iso}(^{1}H)$=0.57 , 0.55mT (#2, #4)and $a_{iso}(^{23}Na)$=0.1mT at g=[2.0078 2.0061 2.0023].

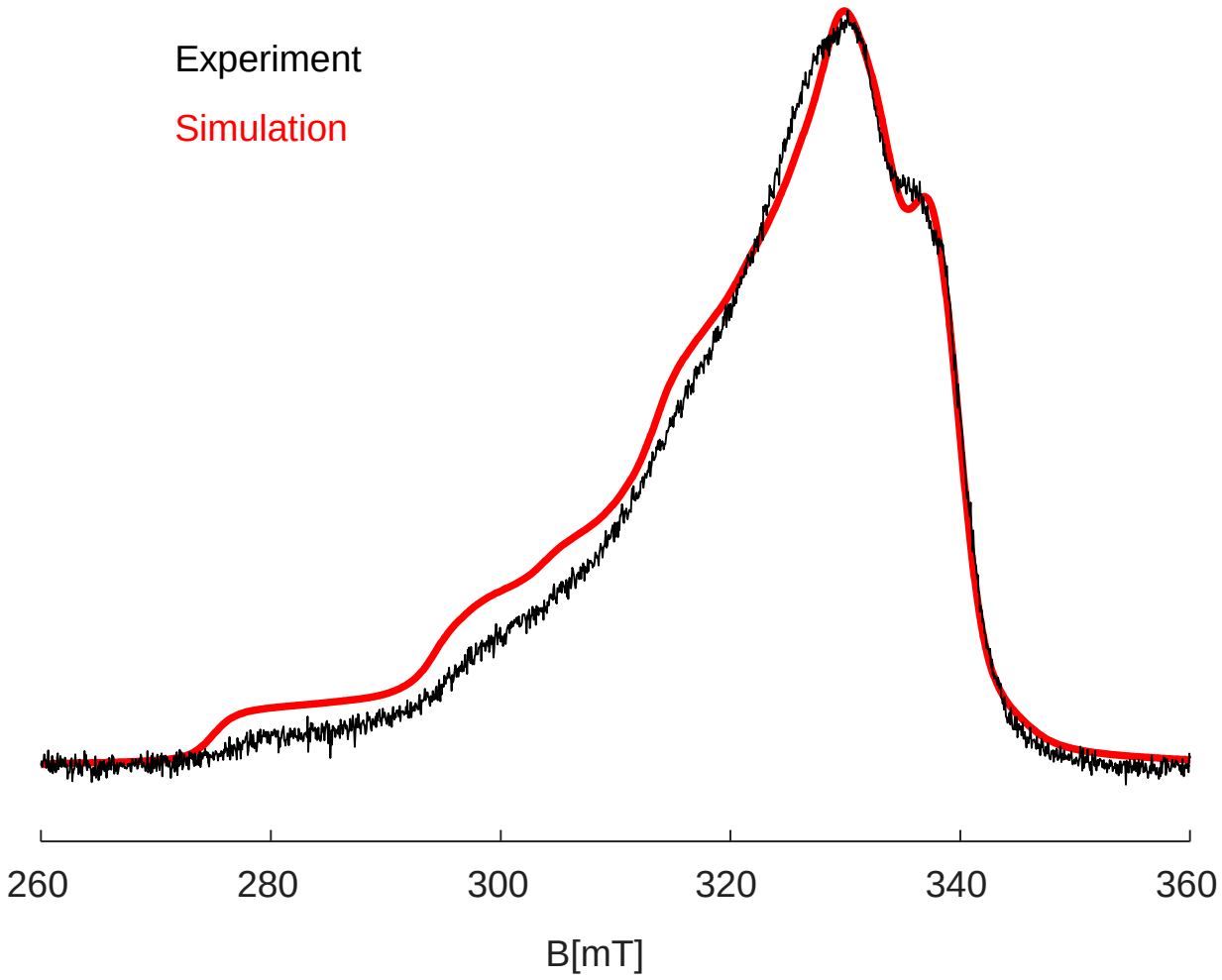



**Figure S5.** Experimental (black) and simulated (red) ESE of Li-DAD in Toluene at 20K at 9.4GHz frequency. Lithium nucleus shows anisotropic hyperfine coupling of A($^{6,7}$Li)=[21 1.7 0.3]mT and two equivalent nitrogens with $a_{iso}$($^{14}$N)=2.5mT at g=[2.192 2.026 1.997].

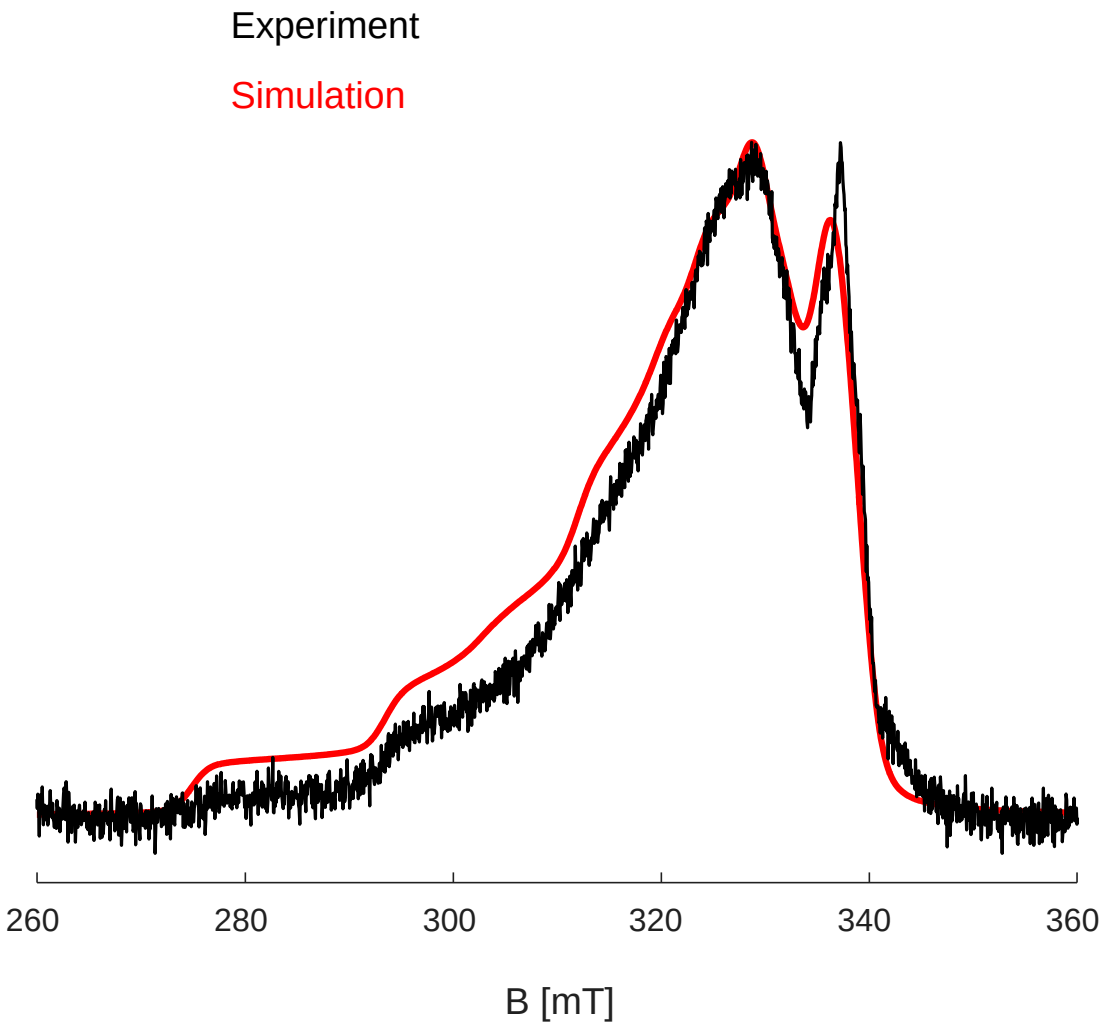
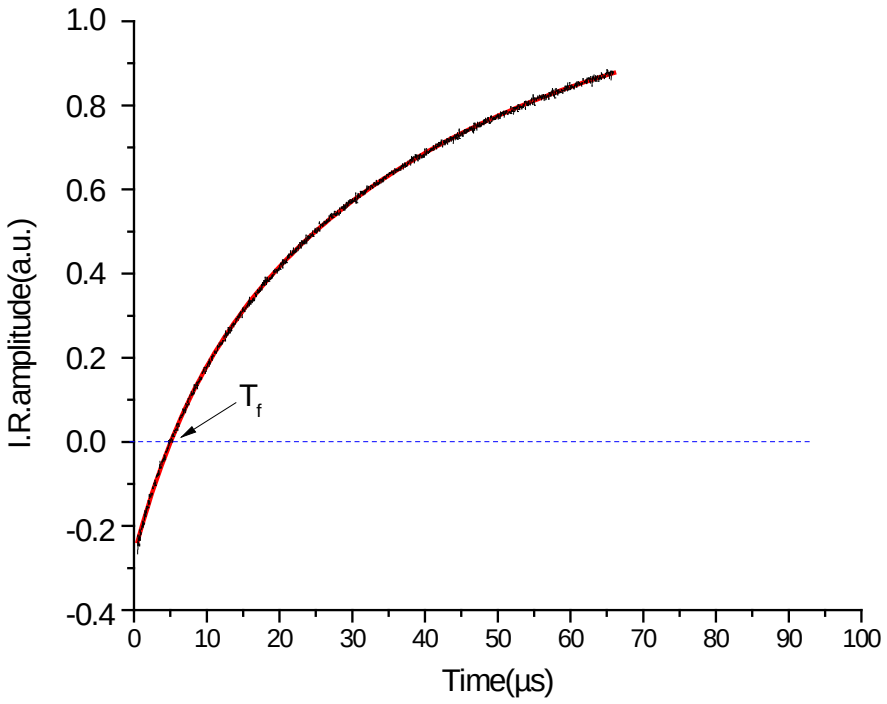
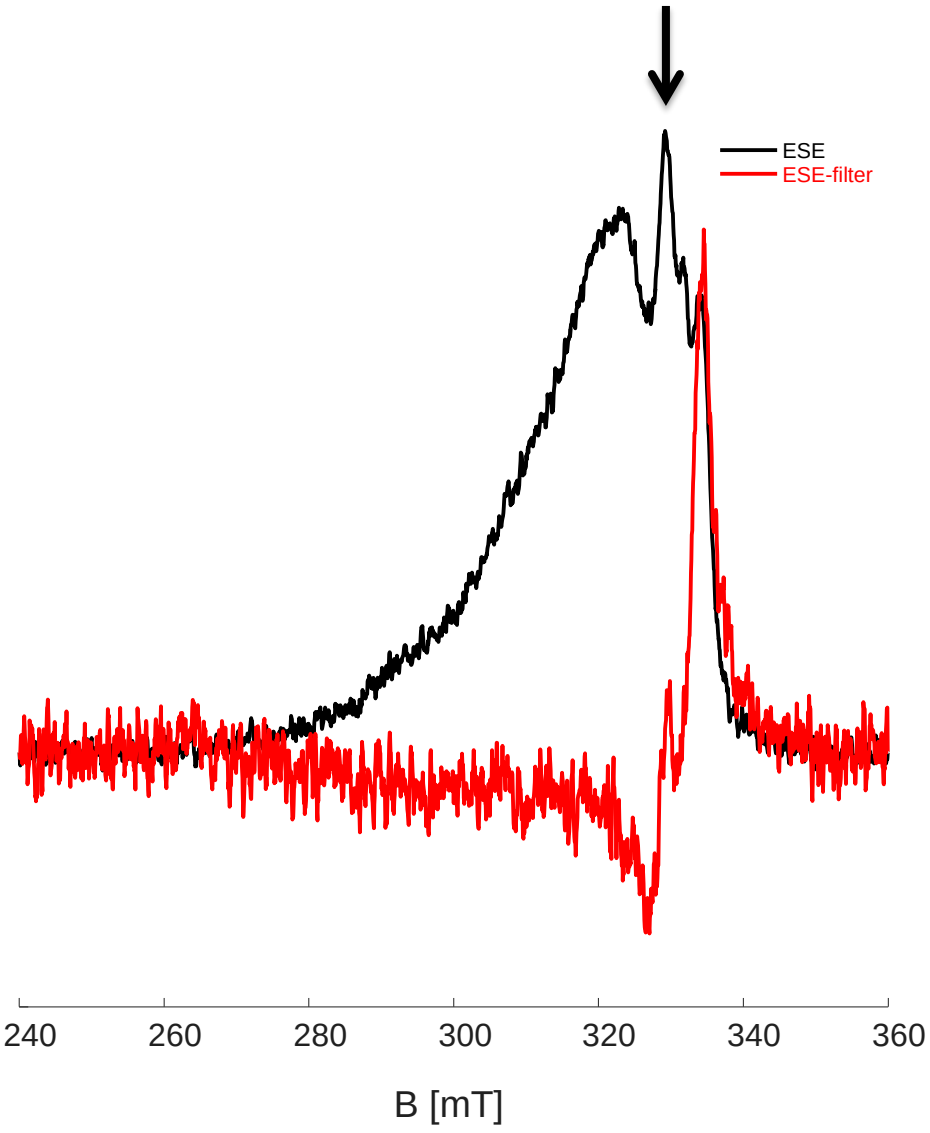



**Figure S6.** Experimental (black) and simulated (red) ESE of Li-DAD in THF at 30K at 9.4GHz frequency. A($^{6,7}$Li)=[20.3 4.2 2.8], A($^{14}$N) =[2.5 1.0 0.3] at g=[2.192 2.026 1.997]. Hyperfine couplings are given in mT.




**FigureS7.** Inversion recovery (IR) trace (black) and its fit (red)recorded for Li-DAD complex dissolved in THF at 20K.




**FigureS8.** Field swept spectra of LiDAD dissolved in THF, recorded before (black) and after(red) applying filter time $T_f$ . The arrow shows the field position for IR trace. All measurements were done at 20K.

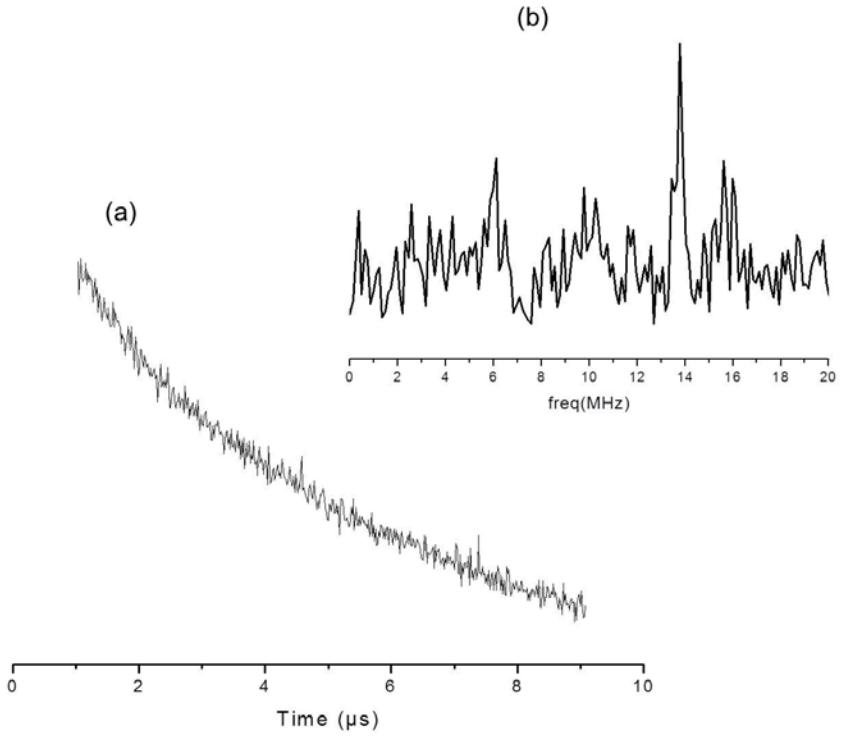
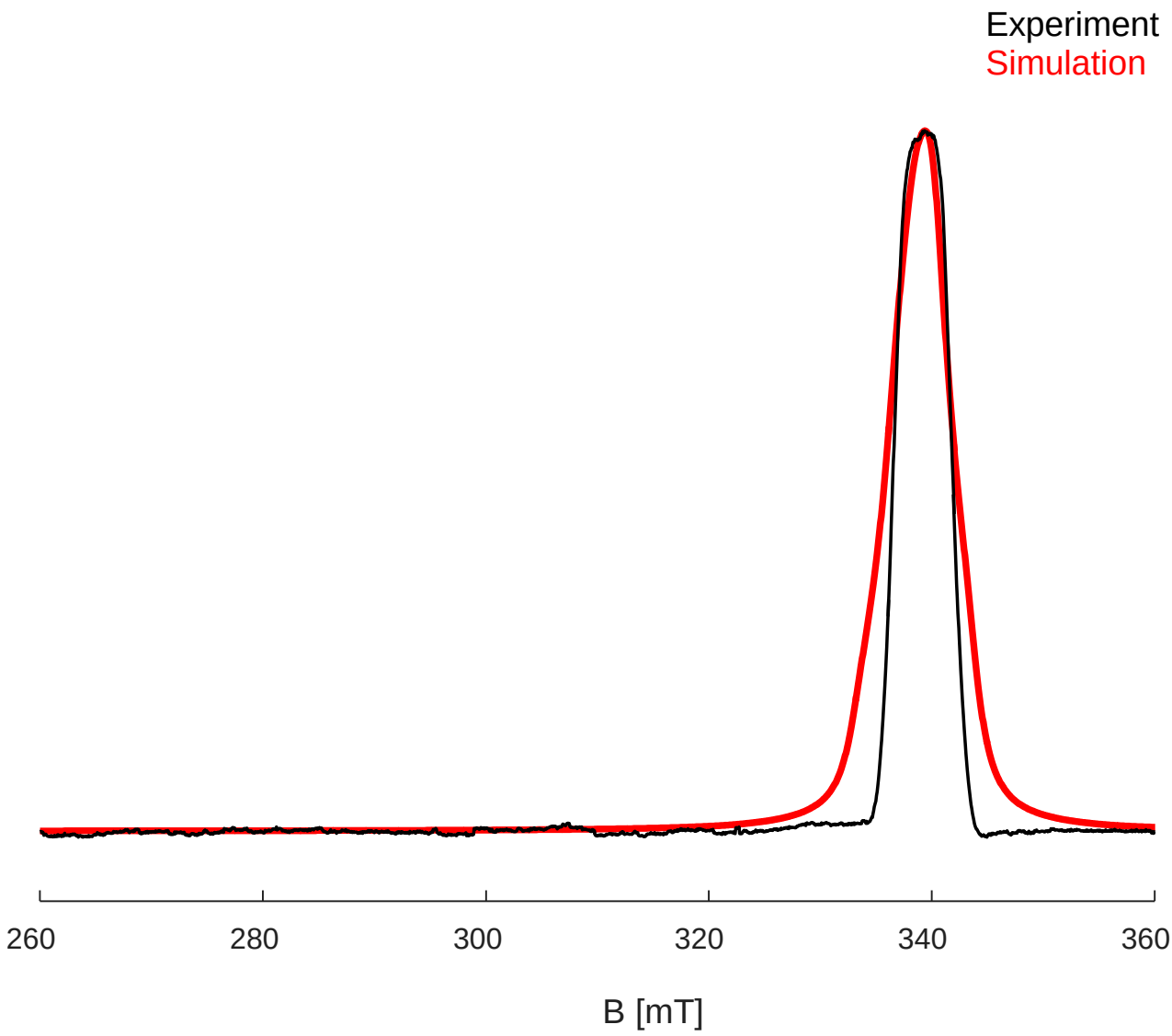
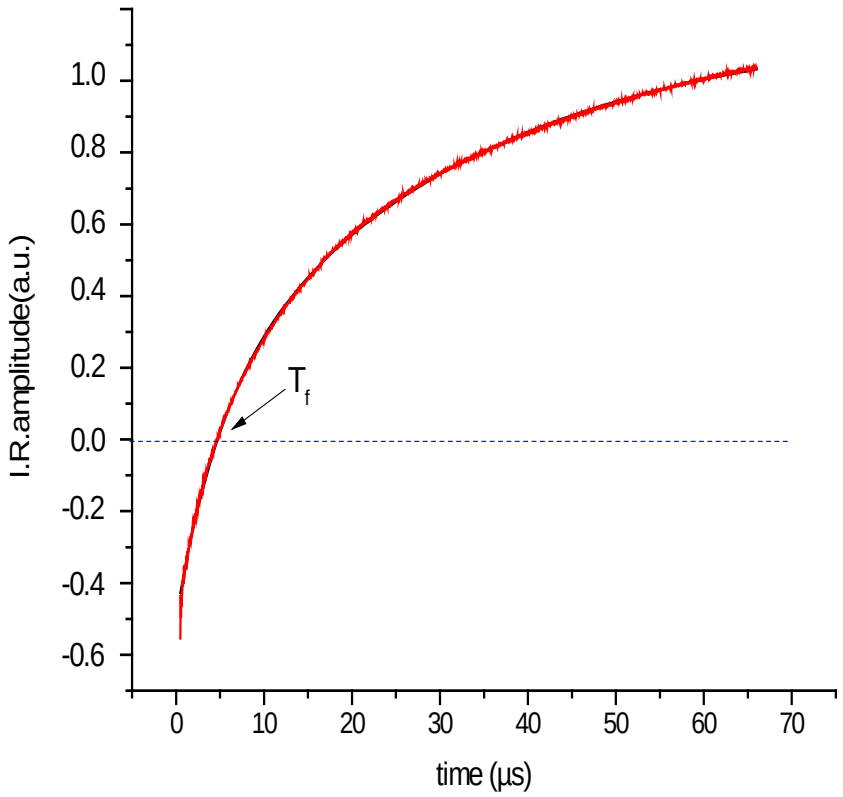



**Figure S9.** Time trace and magnitude spectra of REFINE for Li-DAD in THF at maximum field of 322.8mT. Peaks at 6.1 and13.79MHz could be detected which are in accordance with nuclear Larmor frequency of $^{6,7}$Li and $^{1}$H at resonance field.




**FigureS10.** Experimental (black) and simulated (red) ESE of Na-DAD in THF at 20K at 9.4GHz frequency. A($^{23}$Na)=[3.5 1.7 0.3], A($^{14}$N) =[2.5 1.0 0.35] at g=[2.0089 2.0064, 2.0025]. Hyperfine couplings are given in mT.




**FigureS11.** Inversion recovery (IR) trace (black) and its fit (red) recorded for Na-DAD complex at 20K.

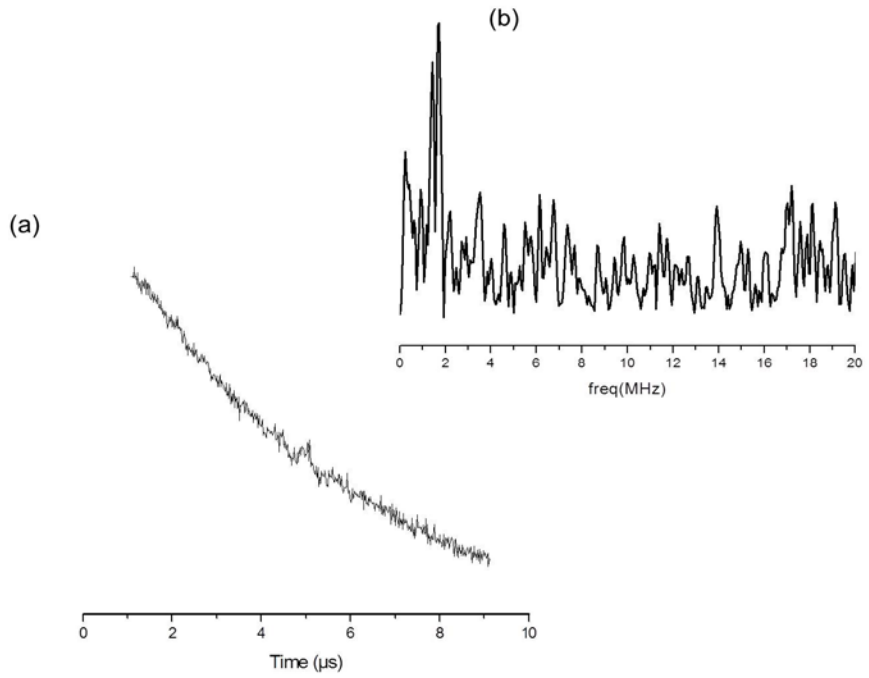



**Figure S12.** Time trace and magnitude spectra of REFINE for Na-DAD in THF at maximum field of 331.8mT, peaks at 1.73, 3.52 and 13.92MHz could be detected which are close to the nuclear Larmor frequency of $^{14}N$, $^{23}Na$ and $^{1}H$ at resonance field.

**TableS1.** Coordinates of the gas phase optimized Li-DAD complex.

| | | | |
|---|---|---|---|
| C | -1.226325 | 0.730622 | -0.571145 |
| C | -2.491151 | 1.460823 | -0.966680 |
| H | -3.390338 | 0.947123 | -0.586983 |
| H | -2.606079 | 1.529983 | -2.064370 |
| H | -2.490704 | 2.484474 | -0.568963 |
| C | -0.139068 | 2.754347 | 0.209446 |
| C | 0.105151 | 3.659059 | -0.857084 |
| C | 0.240088 | 5.023614 | -0.567939 |
| H | 0.437612 | 5.726111 | -1.380676 |
| C | 0.139341 | 5.505783 | 0.733583 |
| H | 0.248189 | 6.573316 | 0.936070 |
| C | -0.092752 | 4.613295 | 1.778054 |
| H | -0.167520 | 4.996519 | 2.797018 |
| C | 0.303306 | 3.169290 | -2.289300 |
| H | 0.012253 | 2.110944 | -2.322147 |
| C | -0.574262 | 3.915229 | -3.307784 |
| H | -0.461651 | 3.468824 | -4.311048 |
| H | -0.297935 | 4.980298 | -3.390000 |
| H | -1.640038 | 3.865938 | -3.029462 |
| C | 1.788699 | 3.236667 | -2.688990 |
| H | 1.940156 | 2.833769 | -3.705876 |
| H | 2.413411 | 2.650938 | -1.994053 |
| H | 2.161217 | 4.275632 | -2.678557 |
| Li | 1.109967 | 0.010039 | 0.385643 |
| N | -0.177582 | 1.358319 | -0.000732 |
| C | -1.140238 | -0.687094 | -0.772225 |
| C | -2.293983 | -1.415415 | -1.426209 |
| H | -2.662647 | -0.878022 | -2.314957 |
| C | 0.129481 | -2.728407 | -0.482316 |
| C | -0.338883 | -3.588644 | 0.545410 |
| C | -0.069644 | -4.960080 | 0.456063 |
| H | -0.422622 | -5.628412 | 1.244662 |
| C | 0.643313 | -5.492465 | -0.614757 |
| H | 0.846273 | -6.564359 | -0.663490 |
| C | 1.099190 | -4.644395 | -1.620647 |
| H | 1.659515 | -5.064945 | -2.458705 |
| C | 0.852812 | -3.266879 | -1.579407 |
| C | -1.092715 | -3.045583 | 1.756799 |
| H | -1.237131 | -1.968069 | 1.596357 |
| C | -2.488139 | -3.674940 | 1.909732 |
| H | -3.031462 | -3.209567 | 2.750381 |
| H | -2.428302 | -4.758388 | 2.111311 |
| H | -3.088665 | -3.536514 | 0.995759 |
| C | -0.272310 | -3.205722 | 3.049603 |
| H | -0.804069 | -2.759029 | 3.907763 |
| H | 0.709507 | -2.710805 | 2.962584 |
| H | -0.088607 | -4.268243 | 3.283932 |
| C | 1.388657 | -2.363977 | -2.686339 |
| H | 0.943590 | -1.370228 | -2.537696 |
| C | 2.916125 | -2.203287 | -2.580891 |
| H | 3.294215 | -1.507266 | -3.350043 |
| H | 3.431026 | -3.170351 | -2.714459 |
| H | 3.210936 | -1.810488 | -1.592210 |
| C | 0.983831 | -2.838112 | -4.091891 |
| H | 1.312784 | -2.109525 | -4.852802 |
| H | -0.110058 | -2.949414 | -4.174547 |

**TableS1.**Contd.

| | | | |
|---|---|---|---|
| N | -0.024434 | -1.325533 | -0.364882 |
| C | -0.233876 | 3.241971 | 1.542184 |
| C | 0.917908 | 1.797584 | 3.261278 |
| H | 0.783583 | 1.030651 | 4.044067 |
| H | 1.560830 | 1.369429 | 2.468730 |
| H | 1.477098 | 2.642598 | 3.699341 |
| C | -1.325237 | 2.806726 | 3.828394 |
| H | -0.842352 | 3.630976 | 4.380120 |
| H | -2.286019 | 3.178539 | 3.437796 |
| H | -1.539589 | 2.007425 | 4.558279 |
| C | -0.438275 | 2.266174 | 2.697667 |
| H | -0.941190 | 1.380353 | 2.282810 |

**TableS2.** Coordinates of the gas phase optimized Na-DAD complex.

| | | | |
|---|---|---|---|
| C | 0.026056 | 0.717398 | 1.147627 |
| C | 0.043810 | 1.440152 | 2.481463 |
| H | 0.828235 | 1.051693 | 3.153616 |
| H | -0.915957 | 1.329335 | 3.020073 |
| H | 0.221393 | 2.515207 | 2.341125 |
| C | 0.095011 | 2.799383 | -0.080550 |
| C | 1.347785 | 3.466994 | -0.159117 |
| C | 1.371274 | 4.851600 | -0.364085 |
| H | 2.332192 | 5.367282 | -0.429950 |
| C | 0.195625 | 5.588369 | -0.485962 |
| H | 0.235264 | 6.667414 | -0.648915 |
| C | -1.030202 | 4.933174 | -0.400414 |
| H | -1.951773 | 5.511814 | -0.496311 |
| C | -1.106248 | 3.550570 | -0.195025 |
| C | 2.658367 | 2.691283 | -0.058942 |
| H | 2.403144 | 1.658159 | 0.214258 |
| C | 3.587610 | 3.236044 | 1.038373 |
| H | 4.479291 | 2.594393 | 1.143077 |
| H | 3.073334 | 3.267959 | 2.013266 |
| H | 3.938801 | 4.257315 | 0.811920 |
| C | 3.383900 | 2.636699 | -1.416098 |
| H | 4.301632 | 2.026764 | -1.348579 |
| H | 3.672485 | 3.644712 | -1.760140 |
| H | 2.739295 | 2.195309 | -2.197115 |
| C | -2.468523 | 2.865800 | -0.131250 |
| H | -2.288588 | 1.813377 | 0.129872 |
| C | -3.377182 | 3.456728 | 0.959395 |
| H | -4.316754 | 2.882592 | 1.032562 |
| H | -3.645861 | 4.506040 | 0.748609 |
| C | -3.171199 | 2.882618 | -1.501333 |
| H | -4.131672 | 2.340002 | -1.459423 |
| H | -2.546161 | 2.406473 | -2.277975 |
| H | -3.379680 | 3.913359 | -1.835653 |
| N | 0.044515 | 1.391587 | -0.016726 |
| Na | -0.000005 | 0.000001 | -1.767957 |
| C | -0.026051 | -0.717398 | 1.147627 |
| C | -0.043799 | -1.440153 | 2.481463 |
| H | -0.828223 | -1.051695 | 3.153618 |
| H | 0.915969 | -1.329336 | 3.020069 |
| H | -0.221382 | -2.515209 | 2.341124 |
| C | -0.095011 | -2.799383 | -0.080552 |
| C | -1.347787 | -3.466992 | -0.159119 |
| C | -1.371277 | -4.851598 | -0.364087 |
| H | -2.332197 | -5.367279 | -0.429953 |
| C | -0.195630 | -5.588368 | -0.485965 |
| H | -0.235269 | -6.667413 | -0.648918 |
| C | 1.030198 | -4.933175 | -0.400417 |
| H | 1.951768 | -5.511815 | -0.496313 |
| C | 1.106246 | -3.550570 | -0.195027 |
| C | -2.658368 | -2.691279 | -0.058943 |
| H | -2.403143 | -1.658156 | 0.214257 |
| C | -3.587611 | -3.236040 | 1.038372 |
| H | -4.479291 | -2.594388 | 1.143077 |
| H | -3.073334 | -3.267957 | 2.013265 |
| H | -3.938804 | -4.257310 | 0.811918 |
| C | -3.383901 | -2.636693 | -1.416098 |
| H | -4.301632 | -2.026757 | -1.348579 |
| H | -3.672488 | -3.644706 | -1.760142 |

**TableS2.** Contd.

| | | | |
|---|---|---|---|
| C | 2.468522 | -2.865803 | -0.131251 |
| H | 2.288588 | -1.813379 | 0.129865 |
| C | 3.377176 | -3.456728 | 0.959399 |
| H | 4.316750 | -2.882594 | 1.032565 |
| H | 3.645853 | -4.506041 | 0.748619 |
| H | 2.882150 | -3.429734 | 1.944305 |
| C | 3.171202 | -2.882630 | -1.501332 |
| H | 4.131676 | -2.340014 | -1.459422 |
| H | 2.546168 | -2.406488 | -2.277979 |
| H | 3.379683 | -3.913373 | -1.835647 |
| N | -0.044515 | -1.391586 | -0.016726 |