Figure S1: Experimental EPR spectrum of spin-labelled lysozyme in the composite (black), and the individual traces of the 4 components simulation shown in figure 2
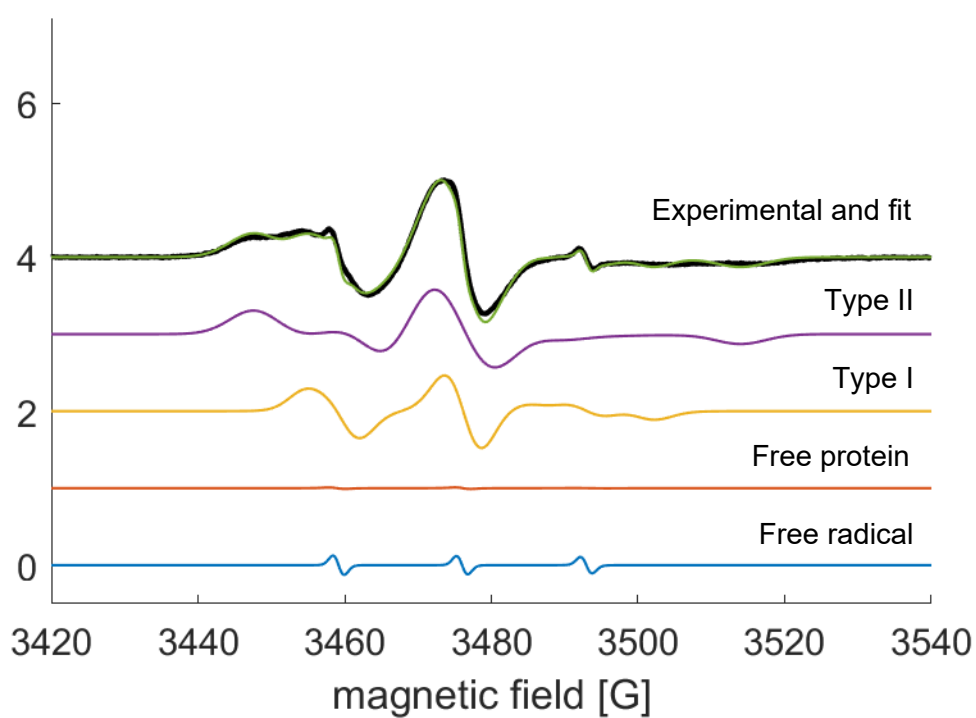
Figure S2: Experimental EPR spectrum of spin-labelled lysozyme in the composite (black), and the individual traces of the 4 components simulation (Table S1). Squared sum of the residuals 1.91

Table S1: Components used for the fit reported in Figure S2.

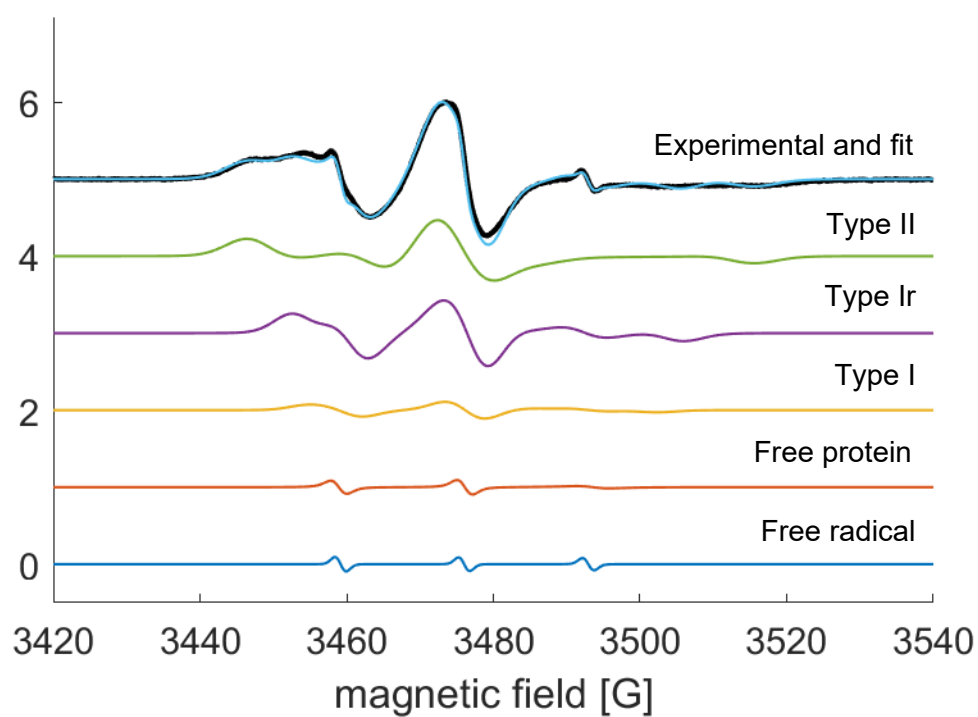| Component	Rotational Diffusion coefficients ($\times 10^6$ s^{-1})	lwpp (G)	L_x (°)	L_y (°)	L_z (°)	weight (%)
Free radical	$4.0 \cdot 10^3$	1.5	0	0	0	0.51
Free protein	22, 28, 12	1.5	62	34	45	0.13
Type I	2.98	4.2	62	34	45	22.1
Type II	0.30	6.7	35	16	2	77.3

Figure S3: Experimental EPR spectrum of spin-labelled lysozyme in the composite (black), and the individual traces of the 5 components simulation shown in figure 2

```
function Tav = quasilibration(T,L)
P = sin(L).*cos(L)./L;
Tav = T;
Tav(1) = Tav(1) + 0.5*(T(3)-T(1))*(1-P(2)) +
0.5*(T(2)-T(1))*(1-P(3));
Tav(2) = Tav(2) + 0.5*(T(3)-T(2))*(1-P(1)) +
0.5*(T(1)-T(2))*(1-P(3));
Tav(3) = Tav(3) + 0.5*(T(1)-T(3))*(1-P(2)) +
0.5*(T(2)-T(3))*(1-P(1));
return
```