Supplementary Information

Figure S1. Perspective drawing of the structure of complex 3 showing the atom numbering. The hydrogen atoms and the acetonitrile molecules are omitted for simplicity.

Figure S2. A view along the $a$ axis showing the hydrogen bonding between the neutral tetranuclear units of complex 3.

Equations connecting the energies of the five calculated states [one high spin and four broken symmetry state] and the exchange parameters:

\[ \begin{align*}
    E_B - E_A &= 8b + 8c \\
    E_C - E_A &= 3a + 4b + 4c \\
    E_D - E_A &= 4b + 4c + 6d \\
    E_E - E_A &= 3a + 8b + 6d
\end{align*} \]

where

$A$, $B$, $C$ and $D$ are the calculated configurations

and

$a = J_{Ni-Ni}$, $b = J_{Cr-Ni}$, $c = J_{Cr-Nia}$ and $d = J_{Cr-Cra}$.