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:

EB – EA = 8b + 8c

EC – EA = 3a + 4b + 4c

ED – EA = 4b + 4c + 6d

EE – EA = 3a + 8b + 6d

where

A, B C and D are the calculated configurations

and

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