

## Supporting Information

### *SI. Crystallographic Details*

Single crystals were mounted on glass fibres, in viscous hydrocarbon oil. Crystal data were collected using a Bruker Apex II diffractometer equipped with monochromated Mo-K $\alpha$  radiation ( $\lambda = 0.71073$  5 Å). All data were collected at 123 K, maintained using an open flow N<sub>2</sub> Oxford Cryostreams cryostat. X-ray data were processed using the SAINT program suite. Structures were solved by direct methods using SHELXS-97<sup>2</sup> and refined by full-matrix least squares against F<sup>2</sup> of all reflections using SHELXL-97<sup>2</sup> with the graphical interface X-Seed.<sup>3</sup> Non-hydrogen atoms were refined anisotropically. Hydrogen atoms were placed in calculated positions and refined using a riding model.

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1 *SAINT*, Bruker AXS Ltd., Wisconsin.

2 *SHELXS-97* and *SHELXL-97*, G.M. Sheldrick, University of Göttingen, 1997.

3 *X-Seed v.2.0*, L.J. Barbour, University of Stellenbosch, 1999.

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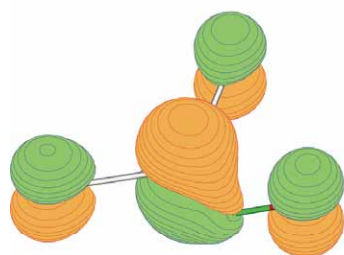
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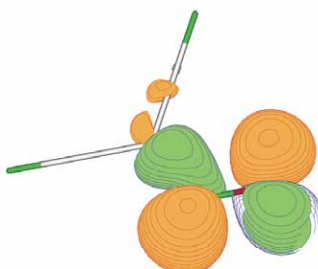
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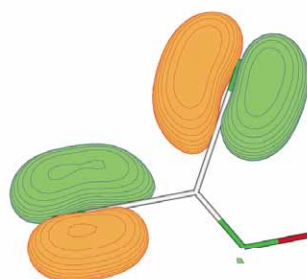
45  
*S2. All calculated HOMOs for the dicyanonitrosomethanide anion, dcnm.*



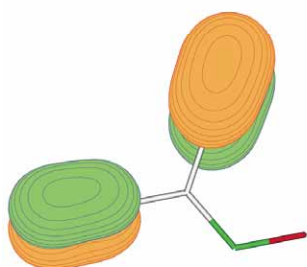
HOMO



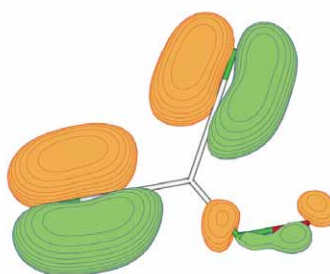
HOMO-1



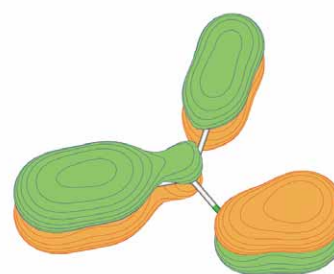
HOMO-2



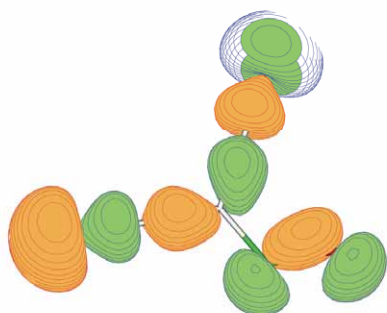
HOMO-3



HOMO-4



HOMO-5



HOMO-6