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Supporting Information

S1. *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-Kα radiation (λ = 0.71073 Å). All data were collected at 123 K, maintained using an open flow N_2 Oxford Cryostreams cryostat. X-ray data were processed using the SAINT program suite. Structures were solved by direct methods using SHELXS-97 [2] and refined by full-matrix least squares against F^2 of all reflections using SHELXL-97 [2] with the graphical interface X-Seed.[3] Non-hydrogen atoms were refined anisotropically. Hydrogen atoms were placed in calculated positions and refined using a riding model.

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.

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