Magnetic Verses Electric-Quadrupole Order in the Mixed Actinide Oxides

Wilkins Stuart B.^{1,2}, Paixão J. A.³, Caciuffo R.⁴, Javorsky P.¹, Wastin F.¹, Rebizant J.¹, Detlefs C.², Bernheoft N.⁵, Lander G. H.¹

¹ European Commission, JRC, Institute for Transuranium Elements, Postfach 2340, Karlsruhe, D-76125 Germany, email: wilkins@esrf.fr

² European Synchrotron Radiation Facility, Boîte Postal 220, F-38043 Grenoble CEDEX, France

³ Physics Department, University of Coimbra, Coimbra, 3004--516 Portugal

⁴ Dipartimento di Fisica ed Ingegneria dei Materiali e del Territorio, Università Politecnica delle Marche, I-60131 Ancona, Italy

⁵ Départment de la Recherche Fondamentale sur la Matière Condensée, CEA, F-38054 Grenoble CEDEX, France

We have used element specific X-ray resonant scattering to investigate the M edge resonances in a single crystal of $U_{0.75}Np_{0.25}O_2$. Earlier neutron diffraction and Mössbauer studies had shown the presence of long-range AF order below $T_N = 16.5$ K, with sizeable magnetic moment both on the U and the Np ions. RXS results confirm the presence of an ordered dipole magnetic moment on the Np ions, but also reveal the presence of AF-electro-quadrupolar on both U and Np ions, with the same propagation vector that defines the magnetic structure. Calculations of the azimuthal dependence for both the Np and U ions using the ATS technique, suggest a 3k transverse structure. A surprising result is that the integrated intensities of Bragg peaks associated with magnetic dipole and electric quadrupole order have different temperature dependences. On cooling, the magnetic dipole order develops first on the uranium ions, with magnetic order on the Np ions following at a lower temperature. At the same temperature, quadrupolar order on both the U and Np ions occurs along with an internal Jahn-Teller lattice distortion.