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Shifts: 12	Local contact(s)**: Danny MANNIX**	

Names and affiliations of applicants (* indicates experimentalists):

Peter NORMILE*, Institute for Transuranium, Karlsruhe

Elizabeth BLACKBURN*, Institute for Transuranium, Karlsruhe

Danny MANNIX*, XMaS C.R.G.

Laurence BOUCHENOIRE*, XMaS C.R.G.

Nicholas BERNHOEFT*, C.E.N.G – DRFMC/SPSMS

Gerard LANDER, Institute for Transuranium, Karlsruhe

Report:

Resonant X-ray scattering (RXS) at the K-edge of As in the two antiferromagnetic (AF) phases of UAs [1] has been studied at the *XMaS* beamline. Working with full polarization analysis, in both AF phases of UAs – 'type-I': *1-k, k* = 1 rlu for 62 K < T < 124 K (= T_N) and 'type-IA': *2-k, k* = 0.5 rlu for 0 < T < 62 K – resonances with asymmetric line-shapes (with respect to the incident photon energy) have been observed for σπ scattering, localised in reciprocal space at the magnetic satellite positions corresponding to each phase. These are shown in Figure 1(a). Previous experimental studies in this field [2,3] did not comment on such asymmetry, but inspection of the (limited) published data (type-IA, only [3]) suggests its presence. Moreover, one observes that the σπ resonances in the type-I and -IA phases have *different* line-shapes, the resonance in the former (high T) phase being less single-Lorentzian-like than that in low-T (type-IA) phase, due to its 'flat-top' peak (note: a log y-scale is used in Fig. 1(a)). The motivation for these measurements was theory [4], which predicted differing line shapes between the two phases, as shown in Figure 1(b). Experimentally, the way in which the line-shapes differ is in disagreement with theory, with the double peak structure in the type-I line-shape (Fig. 1(b)) not being observed (note: the measurement carries a finite energy resolution (~ 2 eV), which will 'smear' such a (calculated) double-peak structure, but not such that it becomes the flat-topped line-shape that has been measured). In addition, the (measured) relative intensities of the resonances in the two magnetic phases disagree strongly with theory: theory puts the ratio of resonant signal strength for type I:type IA phases at ~ 70:1 whereas experimentally the ratio is ~ 1:3.

Also shown in Figure 1(a) is our observation of a signal (resonance) in the σσ channel in the type-IA phase, *only*. This effect was observed at all <00*k*> type satellite positions about the zone centred at (006). The strength of this signal is ~ 10 % of that of the σπ (<00*k*>) resonance in this phase. Given that these K-edge resonance effects are understood to be dipole (E1) in nature, arising from *magnetic-dipole-like* order, a resonance in the σσ channel at <00*k*> (satellite) positions should not be observed [2,4]. The estimated leakage factor of the polarization analyser (PA) is 1/60, thus the origin of the σσ signal (at $1/10^{th}$ of the σπ) cannot be leakage. That it cannot be leakage is also confirmed by the absence of such a (σσ) signal in the type-I phase. The absence of a σσ signal in the type-I phase also implies that πσ scattering, i.e. a 10 % π photon contamination of the incident σ polarized beam, cannot account the σσ (type-IA) signal. (Indeed, the estimated PA leakage factor of 1/60 (given by the intensity ratio, $(006)_{\sigma\pi}/(006)_{\sigma\sigma}$) rules out the possibility of 10 % π photon incident beam contamination.) Finally, we dismiss multiple scattering as the origin, as this ('σσ(IA)') signal was non-vanishing under changes in the sample azimuth (ψ) angle over a range +/- 20 deg.. Due to beamtime limitations, we did not manage to measure the full ψ dependendence (i.e. +/-, at least, 90

deg.) of the σσ(IA) resonance; the UAs sample was initially mounted on the *XMaS* diffractometer with the displex horizontal, and remounted for ψ studies (displex vertical) in the final night of beamtime.

None of the obvious spurious effects, discussed above – those of (i) leakage through the polarization analyzer of the σπ resonance, (ii) non-perfect incident beam polarization and (iii) multiple scattering – can explain the occurrence of the σσ signal at <00*k*> type (satellite) positions in the type IA phase of UAs, with a strength $1/10^{th}$ that of the σπ(IA) signal and concurrent with a complete absence of an equivalent σσ signal in the type I phase. It is clear that more experimental work is required to resolve the origin of this signal. In particular, an experiment should be performed, with the aim of determining the (full) azimuthal-dependence of the signal.

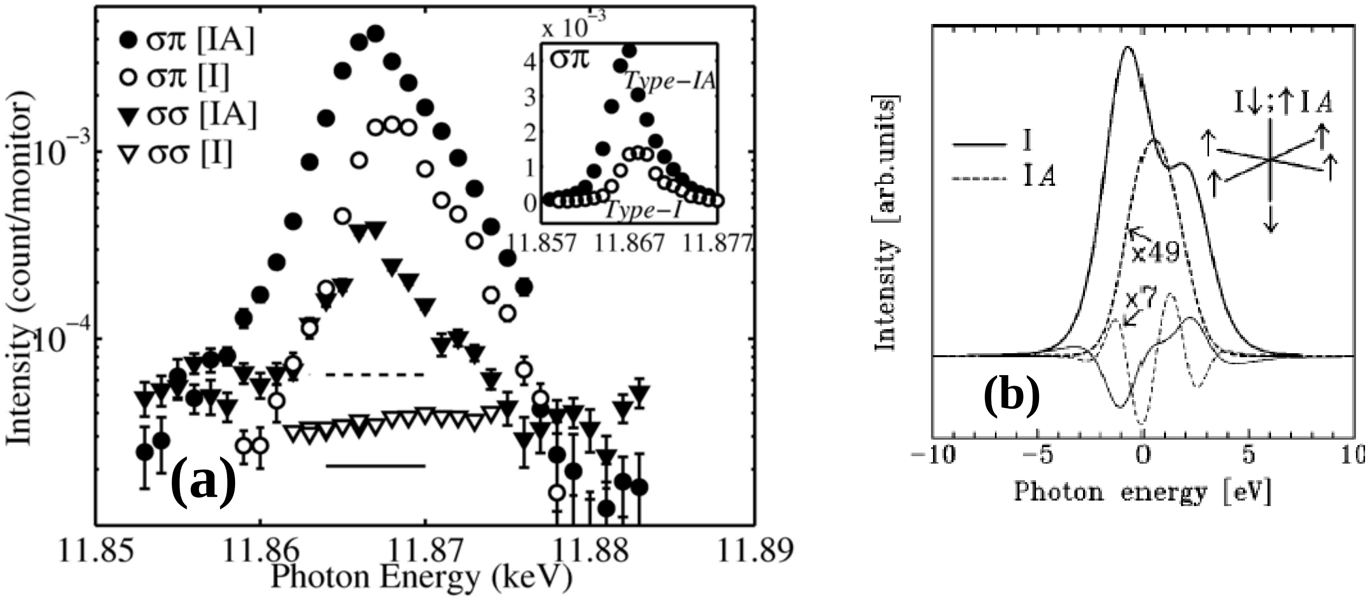

Fig. 1 RXS from UAs in its type-I and –IA, AF phases: **(a)**, experimental (this study; log y-scale) and, **(b)**, theoretical (reproduced from ref. 4; linear y-scale). The experimental intensities (**(a)**) are the results of integrations of sample rocking curve scans through (0 0 6+*k*) type magnetic satellite positions (specifically, the (006.5) and (007) positions for the IA and I, respectively). The dashed and solid lines give, respectively, the estimated leakage into the σσ channels of the σπ signals in the IA and I phases. In **(b),** the solid and dashed lines represent the type I and IA phases, respectively, with the two, larger signals being the (σπ) RXS (the smaller signals are circular dichroism). The inset in **(a)** shows the (measured) σπ signals on a linear y-scale, for comparison with **(b)**.

References and Notes:

[1] The exact sample composition studied was $UAs_{0.975}Se_{0.025}$, this being the only UAs-*like* sample available to us at the time of the study. $UAs_{0.975}Se_{0.025}$ has an identical magnetic phase diagram to UAs, and is thus we referred to as 'UAs'.

[2] Mannix *et al.*, Phys. Rev. Lett. **86**, 4128 (2001)

[3] Longfield *et al.*, Phys. Rev. B **64**, 212407 (2001)

[4] van Veenendaal, Phys. Rev. B **67**, 134112 (2003)