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# Resonant elastic X-ray scattering from 5*f* systems

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**Abstract.** The first REXS experiments on a uranium compound at the U $M_4$ edge (3.728 keV) took place at BNL twenty years ago. An enormous enhancement of the scattering intensity was found. Since that time many other systems have been examined. This paper reviews some of the highlights of resonant scattering from actinide systems, and attempts to extrapolate what might be the future of this field.

Soon after resonant scattering was discovered at the $L$ edges in Ho [1], it was realised that the strongest effects in resonant elastic X-ray scattering (REXS) could be observed if the transitions involve partially occupied shells with strongly spin-polarised states. Thus the largest resonant effects are found when the $L_{2,3}$ edges are used for transition metals of the 3$d$ series, and at $M_{4,5}$ (or $N_{4,5}$) edges for the 4$f$ and 5$f$ series. In the case of the $M_{4,5}$ edges of the actinides (5$f$) the energies are between 3.5 and 5 keV, so within reach of many diffractometers, without the complications of soft X-rays. Hence the first experiment on UAs [2] at BNL in 1989 found the resonant enhancement to be about 6 orders of magnitude.

In the 20 years since this experiment a great many 5$f$ systems have been examined. Some noteworthy highlights are briefly mentioned in this extended abstract.

(1) Extension to *transuranium* materials. At BNL studies were done with NpAs to show both critical scattering and a full examination of the magnetic structure [3,4]. Later, after some difficulties with the crystal surfaces [5], experiments were successfully done on a series of $(U_{1-x}Pu_x)Sb$ single crystals [6,7]. This latter study found an interesting effect at the Pu $M_5$ edge [7], but this has not been confirmed by XMCD experiments, so its origin remains in doubt.

(2) In Ref. [6,7] the *element specific nature* of REXS was well demonstrated. Another study at ID20 (ESRF) examined alloys of $(U_{1-x}Np_x)Ru_2Si_2$, where the parent compound with x = 0 is the famous heavy-fermion "hidden-order" material, which is still not understood. In REXS study [8] scaling gave a magnetic moment of $\sim 0.4\mu_B$ on the U atom. This was attributed to the large molecular field provided by the ordered Np moments (of $1.5\mu_B$). It is now known that the large moment state of $URu_2Si_2$ does indeed have a moment of this magnitude.

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(3) REXS allowed the observation of scattering from the *surface of the antiferromagnet* $UO_2$ [9,10]. These experiments (at BNL) showed that a dead layer was formed at the surface of the crystal, probably due to a change in the stoichiometry, and that the first-order nature of the transition (AF to para) became $2^{nd}$ order at the surface, as expected from theory.

(4) The detection of *higher-order multipoles in 5f systems* was first reported in $UPd_3$ [11,12]. The case of $NpO_2$ is extraordinary, as there is *no* dipole moment, but the quadrupolar moment is induced [13] as a consequence of the ordering of the trikontadipole (rank 5) [14]. This has not been observed directly, but was inferred from neutron inelastic scattering experiments at the ILL [15]. Quadrupolar ordering was also observed directly in $UO_2$ [16], and in a mixed $UO_2$–$NpO_2$ system [17].

(5) The detection of *strong signals at the anion K edges* was surprising [18], and is a consequence of the hybridization between the actinide $5f$ and anion $p$ states [19]. The effect was demonstrated in $UGa_3$ and UAs [18], and also in $UTGa_5$ [20] and $NpTGa_5$ [21] systems.

(6) The use of the *coherent aspect of a synchrotron beam* can be illustrated with *speckle* experiments, which have been attempted on UAs at the U $M_4$ edge [22], but not yet fully exploited. The coherence of the beam also plays a role in defining the energy widths in situations where the absorption length is shorter than the probe coherence length [23].

(7) Finally, exploiting the large intensities at the U $M_4$ resonance we have observed *additional reflections in multi-**k** structures* that are not anticipated from straightforward diffraction theory [24]. Extra reflections appear in positions corresponding to the notation $\langle kkk \rangle$, where the primary magnetic ordering is characterised by $\langle k00 \rangle$. These reflections have an intensity some $10^{-4}$ of that of the primary AF reflections. After a considerable effort they have also been observed by neutron diffraction at the ILL [25]. They clearly are a direct consequence of the multi-**k** magnetic arrangement, but why they appear, and what exactly is their structure factor, are still unclear.

## 1 Conclusions and future prospects

Much remains to be done. Many areas have just been touched upon, e.g. surface effects in antiferromagnets [9], speckle and use of coherent beams [22,23], and can be further exploited. This can be of wider interest with the examination of interface magnetism. We have started down this road with experiments involving multilayers, and recently showed how the off-specular resonant reflectivity can give information about the strong pinning of Gd moments in a U/Gd multilayer [26].

Epitaxial films of uranium have been made [27,28], and more recently of $UO_2$ (unpublished). There is no magnetism in uranium, but REXS may be able to help with understanding the interfacial features – up to now efforts have been concentrated on resonant reflectivity and understanding how the induced moments (by hybridisation with neighbouring Fe layers) are distributed within the U layers of the multilayers [29]. In this respect XMCD measurements, another element-specific technique, are also very valuable [30].

In the case of $UO_2$ the production of thin films will allow further studies by REXS of the antiferromagnetism in such samples. Perhaps of greater general interest, these films will also allow the study of chemical changes at the surface of $UO_2$ with electrochemistry. These will be performed with grazing incident techniques, but the greater sensitivity to uranium if one works at the $L_3$ absorption edge gives an additional motivation for REXS.

In general, if epitaxial films can be produced of actinide systems, then a great deal of new science can be performed. Surface scattering experiments looking at truncation rods can benefit from using the $L_3$ edges for charge effects, and the $M$ (or $N$) edges for magnetism. Of particular interest, for example, would be to study interfacial effects involving actinides by using the technique of examining the crystal-truncation rods. Such techniques could not only bring new information to the problem of actinide migration, but also to fundamental physics involving heavy-fermion systems that are part of larger heterostructures. The small quantities (micrograms) needed means that it is possible to envisage experiments right up to Cm in the actinide series. Of course, such samples would have to be prepared and capped in a special laboratory, but, once prepared, their activity would be low enough that experiments could certainly be performed at the ESRF. Perhaps one of the most interesting experiments would be on curium metal. It is known to be antiferromagnetic at $\sim$60 K, but the form of the antiferromagnetism is unknown. Given that the coupling is *different* for Cm than for Gd (which lies below Cm in the $4f$ series), the nature of the antiferromagnetic arrangement is of considerable interest, as a small orbital moment should exist in Cm [31].

It is unfortunate that at this time the ESRF has closed the principal beamline (ID20) involved in these studies, but the work continues at other beamlines and synchrotrons.

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