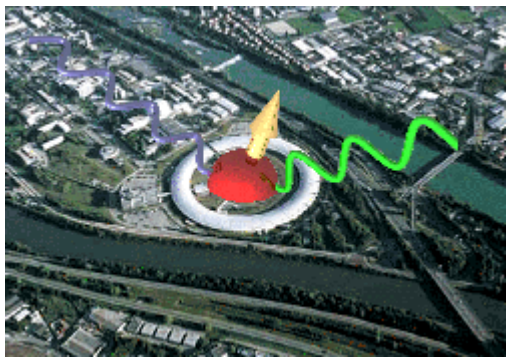
International Workshop on Resonant X-ray Scattering in Electrically-ordered Systems

last modified 25-09-2006 10:12



Dates:	12 - 13 February 2004
Venue:	ESRF, Auditorium
Organisers:	Luigi Paolasini, Carsten Detlefs, Simon Brown
Workshop Secretary:	Claudine Brun

Scope of the Workshop

This Workshop will be held at the ESRF, Grenoble (France) **from Thursday 12th to Friday 13th February 2004.**

The research field of Resonant X-ray Scattering (RXS) has achieved tremendous progress in the last years. Nowadays RXS is rapidly becoming the crucial technique for investigating the subtleties of microscopic magnetism in systems where the ground state properties reflect a delicate balance between several different correlated processes.

The aim of this workshop is to discuss **present and future possibilities for RXS investigations of electronic order**, including studies of charge, magnetic, and multipolar ordered states. The sessions will cover experimental and theoretical aspects of hard and soft X-ray resonant scattering from single crystals and thin films. The workshop will feature invited lectures and shorter talks of 15 min. (incl. discussion). **A poster session** in the afternoon on the first day should foster intensive discussion among the participants.

International Workshop
on Resonant X-ray Scattering in Electrically-Ordered Systems

12-13th February 2004

PROGRAMME

Venue: ESRF Auditorium

Thursday 12th February 2004

08:30 Registration

09:00 Welcome by **Francesco SETTE,** *Director of Research ESRF*

Session I — **Christian VETTIER**

09:15 **Steve COLLINS** - *Diamond Light Source Didcot, UK*
Identification of Quadrupole Transitions at Absorption Edges

09:40 **Kohtaro ISHIDA** - *Tokyo University of Science, Japan*
Resonant and Non-Resonant Scattering Observed in the Forbidden Reflections of Hematite (Fe203) and Eskolaite (Cr203)

10:05 **Carlos GILES** - *University of Campinas, Brazil*
Probing the Helical Quadrupolar Order in Ho with Resonant and Non-Resonant X-ray Scattering

10:30 ***Coffee Break***

11:00 **Changyong SONG** - *POSTECH Pohang, South Korea*
Interference of Magnetic and ATS Reflections in Resonant X-ray Scattering of GdB4

11:25 **Stuart WILKINS** - *Institute of Transuranium Elements JRC Karlsruhe, Germany*
Magnetic Verses Electric-Quadrupole Order in the Mixed Actinide Oxides

11:40 **Alessandro BOMBARDI** - *ESRF Grenoble, France*
Direct Determination of the Magnetic Ground State in the Square Lattice S=1/2 Antiferromagnet Li_2VOSiO_4

12:00 ***Lunch at the ESRF/ILL restaurant***

Session II — **Bill STIRLING**

14:00 **Jonathan GOFF** - *University of Liverpool, UK*
Resonant X-ray Scattering from Magnetic Multilayers

14:25 **Emilio LORENZO DIAZ** - *Laboratoire de Cristallographie CNRS Grenoble, France*
Charge Ordering as seen by RXS

14:40 **Urs STAUB** - *Swiss Light Source at Paul Scherrer Institute Villigen, Switzerland*
Charge Ordering at Metal-Insulator Transitions Studied by X-rays

15:10 ***Coffee Break***

15:40 **George SRAJER** - *Argonne National Laboratory, USA*
Inhomogeneous Magnetic Phase Transition in Fe/Gd Multilayers

16:05 **Laurence BOUCHENOIRE** - *ESRF Grenoble, France*
Resonant X-ray Scattering of U/Fe Multilayers at XMaS Beamline

16:20 **Gloria SUBIAS PERUGA** - *Instituto de Ciencia de Materiales de Aragon Zaragoza, Spain*
Structural Model for the X-ray Resonant Scattering: Half-Doped Manganites and Magnetite

17:00 ***Poster Session and Cocktail***

19:30 ***Workshop Dinner at the restaurant 'Le Pudding', Le Sappey en Chartreuse***

International Workshop
on Resonant X-ray Scattering Electrically-Ordered Systems

12-13th February 2004

PROGRAMME

Venue: ESRF Auditorium

Friday 13th February 2004

Session III — **Calogero NATOLI**

09:00 **Stephen LOVESEY** - *Diamond Light Source Didcot, UK*
A Case for Np 5f Octupole and Hexadecapole Motifs in NpO_2

09:25 **Vladimir DMITRIENKO** - *A.V. Shubnikov Institute of Crystallography Moscow, Russia*
Atomic Vibration Effects in the Resonant 'Forbidden' Reflections

09:50 **Paul STRANGE**- *Keele University, UK*
Ab-Initio Theory of Resonant X-Ray Scattering

10:15 ***Coffee Break***

10:45 **Jun-Ichi IGARASHI** - *JAERI Hyogo, Japan*
Resonant Magnetic X-ray Scattering from Transition-Metal Compounds and Actinide Compounds

11:10 **Sergio DI MATTEO** - *Laboratorio Nazionale di Frascati, Italy*
Resonant X-ray Scattering as a Tool to Detect Parity-Odd Magnetic Multipoles in Globally Centro-Symmetric Crystals

11:25 **Nicholas BERNHOEFT** - *CEA Grenoble, France*
The Unresolved Origin of <kkk> Reflections in UAs0.8Se0.2

12:00 ***Lunch at the ESRF/ILL restaurant***

Session IV — **Doon GIBBS**

14:00 **John HILL** - *Brookhaven National Laboratory, USA*
Ground States and Excitations in Half-Doped Manganites: Hard and Soft X-ray Scattering Studies

14:25 **Jonathan LANG** - *Argonne National Laboratory, USA*
Imaging Spiral Magnetic Domains in Rare Earth Metals

14:50 **Peter HATTON** - *University of Durham, UK*
Soft X-ray Resonant Diffraction from Single Crystals

15:15 ***Coffee Break***

15:45 **Koichi KATSUMATA** - *SPring8/RIKEN Hyogo, Japan*
Synchrotron X-ray Diffraction Measurements in High Magnetic Fields and Low Temperatures

16:10 **Daniel BRAITHWAITE** - *CEA Grenoble, France*
High Pressure Resonant Magnetic X-ray Scattering: Dream or Reality?

16:30 ***Final Discussion and Conclusions***

17:00 ***End of the Workshop***

Identification of Quadrupole Transitions at Absorption Edges

Collins S P[1], Pettifer R F[2] and Laundy D[3]

[1] Diamond Light Source Ltd, UK, email: steve.collins@diamond.ac.uk
[2] University of Warwick, UK
[3] Daresbury Laboratory, UK

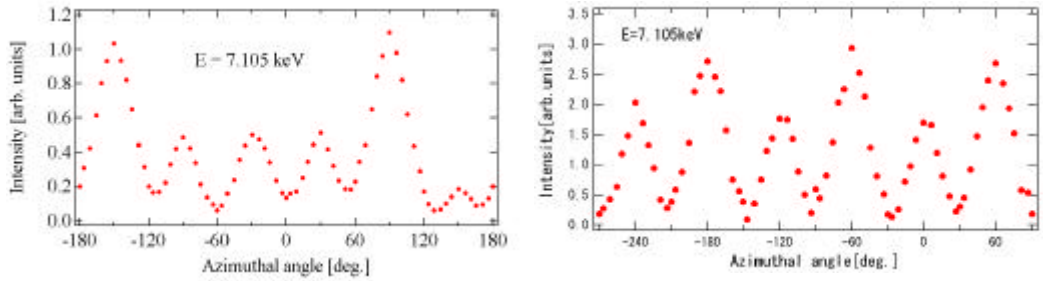
In recent years, several techniques have been put forward to identify electric quadrupole transitions in x-ray absorption spectroscopy and scattering. Such transitions are of fundamental interest, and allow, via the quadrupole selection-rules, sensitivity to valence states that are typically accessible only via soft x-ray spectroscopy. This talk describes some recent experimental results from the SRS, Daresbury Laboratory, in which pre-edge quadrupole features in Gd3Ga5O12 have been strongly enhanced, allowing unambiguous identification. Future possibilities for studies using this new approach will be outlines.

Resonant and Non-Resonant Scattering Observed in the Forbidden Reflections of Hematite(Fe_2O_3) and Eskolait(Cr_2O_3)

Ishida K. and Kokubun J.

Faculty of Science and Technology, Tokyo University of Science, Noda, Chiba 278-8510, Japan
email: ishida@ph.noda.tus.ac.jp

Both hematite and eskolait have corundum structure and are antiferromagnetic at room temperature. The direction of magnetic moment is nearly in the c-planes. In hematite, the moment of adjoin planes are oposit direction but the moment of a c-plene is zero in eskolait. Therefore, X-ray magnetic secattering of 00*l* reflections can be observed in hematite [1] but not in eskolait. Near the Fe K-absorption edge, Finkelstein et al. measured resonant scattering of the "forbidden" 003 reflection in hematete and observed that the azimuthal angle dependence of the intensity was six-fold symmetry.[2] We measured the resonant scattering of the "forbidden" 003 and 009 reflections near the Fe (Cr) K-edge in hematete (eskolait).[3] In hematite the energy spectrum shows non-resonant intensity as well as resonant one. But, there is only resonant scattering in eskolait. Azimuth depentence of the resonant 003 reflection in hematite shows three-fold symmetry but in eskolait, six-fold one. The six-fold symmetry agrees with the calculation based on quadrupole transition. The three-fold symmetry can be explaned by interference between dipole-quadrupole and quadrupole scattering. For non-resonant intensity in hematite, the azimuth dependence shows two-fold symmetry. Below the Morin temperature (T_M=250K), the direction of magnetic moment changes parallel to the c-axis. The observed non-resonant intensity disappeared below the T_M. The temperature dependence is consistent with that of neutron measurements for the 003 reflection. Therefore, the non-resonant intensity is due to magnetic scattering caused by the antiferromagnetic structure. For the 009 reflection, azimuthal dependence in hematite shows rather complicated behavior, neither three-fold nor six-fold symmetry as shown in Fig. 1 (a). But in eskolait, it shows six-fold symmetry. Azimuth dependence of the 009 in hematite becomes three-fold symmetry below the Morin temperature as shown in Fig. 1 (b). This means some contributions of magnetic effect to the electric resonant scattering.



(a) (b)

Figure 1: Azimuth dependence of the 009 resonant reflection in hematite observed at (a) room temperature and (b) 150 K

References

[1] – F.de Bergevin and M. Brunel, Acta Crystallogr. A **37**, 324, (1981)
[2] – K.D. Finkelstein, QunShen and S. Shastri, Phys. Rev. Lett. **69**, 1612, (1992)
[3] – J. Kokubun and K. Ishida, Photon Factory Activity Report 2002 #**20**, 21 (2003)

Probing the Helical Quadrupolar Order in Ho with Resonant and Non-Resonant X-ray Scattering

Giles C., Yokaichiya F.

Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, C.P. 6165, 13083-970 Campinas-SP, Brazil, email: giles@ifi.unicamp.br

The 4f aspherical charge density satellite peaks in Ho measured by X-ray scattering, initially studied by Keating [1], were investigated both in the spiral antiferromagnetic and ferromagnetic phases at the Brazilian Synchrotron Light Laboratory (LNLS) [2]. Temperature dependences of the 2τ satellite in the planar spiral phase and the τ satellite in the conical phases show the same dependence as the magnetic ordering [3]. Q-dependence of the even satellites evidences the quadrupolar nature of the satellites and is in good agreement with theoretical calculations using the formalism for multipolar scattering with the Stevens equivalent-operator method [4]. Unexpected integrated intensities for the even satellites indicate the effect of the anisotropy of the tensor susceptibility (ATS) on these incommensurately modulated ordering peaks. Resonant X-ray scattering of the satellites at the L_3 edge were also investigated giving indication of interference effects with the extra magnetic satellite peaks appearing at resonance [5].

References

[1] - D. T. Keating, Phys. Rev. **178**, 732, (1969)
[2] - C. Giles, F. Yokaichiya, S. W. Kycia, L. C. Sampaio, D. C. Ardiles-Saravia, M. K. K. Franco, R. T. Neuenschwander, J. Synchrotron Rad. **10**, 430, (2003)
[3] - F. Yokaichiya, C. Giles, Physica B, (2004) in press
[4] - H. Westfahl, (2003). Unpublished
[5] - D. Gibbs, D. R. Harshman, E. D. Isaacs, D. B. McWhan, D. Mills, C. Vettier, Phys. Rev. Lett. **61**, 1241 (1988); J. P. Hannon, G. T. Trammel, M. Blume, D. Gibbs, Phys. Rev. Lett. **61**, 1245 (1988)

Interference of Magnetic and ATS Reflections in Resonant X-ray Scattering of GdB_4

Song C.[1], Lee K.-B.[1,2], Ji S.[1], Koo J.[1], Park ,Y.-J.[1,2], Kim J.-Y.[2], Park J.-H.[1], Shin H.J.[2], Rhyee J. S.[3], Oh B. H.[3], and Cho B. K.[3]

[1]electron Spin Science Center and Physics Department, Pohang University of Science and Technology, Pohang, 790-784, Korea, email: cysong@postech.ac.kr
[2]Pohang Accelerator Laboratory, POSTECH, Pohang, 790-784, Korea
[3]Department of Materials Science and Engineering, K-JIST, Kwangju, 500-712, Korea

Resonant X-ray scattering experiments at the Gd L_3 absorption edge show interference between magnetic and anisotropic tensor susceptibility (ATS) reflections in GdB_4. The interference behavior is manifested with the phase shift, **? Q,?),** in the azimthal rotation (y ||**Q**) of the (2n+1 0 0) superstructure reflections; I~cos^2[y –D(**Q,** x)**]**. The phase shift depends on reflection planes (**Q**) and the relative strength (x) of magnetic and ATS scattering.

Further, the energy profiles are obtained from the magnetic and ATS resonances exihibiting ~10 eV separation between the maximum resonance energies. The findings show that the Gd 5*d* band experienced hybridization giving rise to a significant split into isotropic lower energy band and distorted upper band states that account for the magnetic and ATS scattering, respectively.

References

[1] - S Ji, C. Song, J. Koo, K.-B. Lee, Y. J. Park, J. Y. Kim, J.-H. Park, H. J. Shin, J. S. Rhyee, B. H. Oh, and B. K. Cho, Phys. Rev. Lett., **91**, 257205 (2003).

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 3**k** 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.

Determination of the Magnetic Ground State in the Square Lattice S=1/2 Antiferromagnet Li_2VOSiO_4

Bombardi A.[1], de Bergevin F.[1], di Matteo S.[2], Paolasini L.[1], Rodriguez-Carvajal J.[3], Carretta P.[4], Millet P.[5], and Caciuffo R.[6]

[1]European Synchrotron Radiation Facility, B.P. 220, 38043 Grenoble Cedex 9, France, e-mail bombardi@esrf.fr
[2]Udr Roma III, via della Vasca Navale 142, 00100 Roma, Italy
[3]LLB, CEA- Saclay, 91191 Gif sur Yvette Cedex, France
[4]Dipartimento di Fisica, Università di Pavia, Via Bassi 6, I-27100 Pavia, Italy
[5]Centre d'Elaboration des Matériaux et d'Etude Structurales, CNRS, 31055 Toulouse Cedex France
[6]Dipartimento di Fisica e Ingegneria dei Materiali, Università Politecnica delle Marche, Via Brecce Bianche, I 60131 Ancona, Italy

Powder neutron diffraction and resonant X-ray scattering measurements from a single crystal have been performed to study the low-temperature state of the 2D frustrated, quantum-Heisenberg system Li_2VOSiO_4.

Both techniques indicate a collinear antiferromagnetic ground state, with propagation vector k=(1/2 1/2 0), and magnetic moments in the a-b plane. Contrary to previous reports, the ordered moment at 1.44 K, m= 0.62(3) μ_B, is very close to the value expected for the square-lattice Heisenberg model (~ 0.6 μ_B). The magnetic order is three dimensional, with antiferromagnetic a-b layers stacked ferromagnetically along the c-axis. Neither X-ray nor neutron diffraction show evidence for a structural distortion between 1.6 and 10 K.

Resonant X-ray Scattering from Magnetic Multilayers

Goff J.P.[1], Deen P.P.[1], Lee S.[1], Stunault A.[2], Brown S.[2], Mannix D.[2], McIntyre G.J.[3], Ward R.C.C.[4] and Wells M.R.[4]

[1] Department of Physics, University of Liverpool, Oliver Lodge Laboratory, Liverpool L69 7ZE, United Kingdom, email: jpgoff@liverpool.ac.uk
[2] European Synchrotron Radiation Facility, BP 220, F-38043, Grenoble Cedex 9, France
[3] Institut Laue-Langevin, BP 156, F-38042, Grenoble Cedex 9, France
[4] Oxford Physics, Clarendon Laboratory, Oxford OX1 3PU, United Kingdom

In this talk I shall describe two ways in which resonant X-ray scattering on XMaS has provided new understanding of the properties of magnetic multilayers.

Previous attempts to study induced spin-density waves in the spacer layers of multilayers using resonant X-ray scattering had been unsuccessful, but this was achieved for the first time on ID20 using Nd/Pr multilayers, where it was possible to investigate the magnetism of the Nd and Pr components separately. Energy scans revealed strong magnetic resonances at the Nd and Pr L_{II} edges, but with shoulders at higher energies. I shall describe how measurements on XMaS have allowed us to understand the anomalous line shape at these edges. The wave-vector transfer dependence of the scattering shows that both components of this resonance are due to dipolar transitions from the *2p* core levels to the *5d* bands. This clearly demonstrates that we are directly measuring the conduction-electron spin-density wave responsible for the propagation of magnetic order. The structure of the L_{II} resonance is believed to arise because the width of the *5d* band is broad in comparison to the inverse core hole lifetime.

The interactions between the ferromagnetic and antiferromagnetic blocks in Fe/Mn multilayers are of great technological interest since such interactions are responsible for the exchange biasing in spin valves. Theory predicts that the Mn should form either an antiferromagnetic structure with spins in the same direction as the Fe, or a helical structure. The extraordinary result obtained using neutron diffraction is that the Mn orders with a simple antiferromagnetic structure, but with spins pointing along the growth direction perpendicular to those of Fe. We have studied the interfacial structures using anomalous X-ray diffraction on XMaS. Fe/Mn multilayers have a one-electron contrast and the diffraction harmonics are, therefore, very sensitive to the small changes to the scattering factors at the Fe and Mn K edges. The results show that the interfaces contain terraces, leading to a frustration in the exchange interaction between Fe and Mn that can explain the observed magnetic ordering in the Mn.

Charge Ordering as seen by RXS

Lorenzo J.E., Joly Y. and Nazarenko E.

Laboratoire de Cristallographie, CNRS, BP-166X, 38042 Grenoble Cedex 09,
email: lorenzo@grenoble.cnrs.fr

Electronic orderings in the form of charge and orbital ordering are certainly one of the most intriguing subjects in contemporary solid state physics. There are just a handful of experimental techniques which are directly sensitive to the microscopic properties of these elementary orderings and resonant X-ray scattering (RXS) is certainly one of the best suited techniques. In this paper we will discuss charge ordering issues revealed through (i) the analysis of our RXS data on NaV_2O_5 and Fe_3O_4., (ii) *ab-initio* calculations of the observed spectra and (iii) analysis of the symmetry of the structure factor tensors. Finally, and in view of the rather large amount of contradictory results issued from the resonant scattering technique, we will try to shed light on the data analysis by pointing out what can (and can not) be measured with this technique.

Charge Ordering at Metal-Insulator Transitions Studied by X-rays

Staub U.

Swiss Light Source, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland, email: Urs.Staub@psi.ch

Two examples of resonant X-ray scattering for the study of charge order on materials exhibiting a “metal-insulator transition is given. The first, the exotic f-electron material Yb_4As_3, where the charge order is incomplete and the resonant X-ray scattering (RXS) data can be easier interpreted due to the well-predicted energy dependence of the scattering factors. The RXS results are compared with bond valence sum considerations and the non-resonant X-rays scattering data. The second, the $RNiO_3$ perovskites, where resonant X-ray scattering is able to extract a quantitative charge difference of the two Ni sites in the insulating phase. Even though azimuthal angle dependence for the integrated intensities of the charge reflections is observed, it can be shown that this is not and indication of a simultaneous occurring of orbital ordering but only a unique property of the strain in the epitaxial film used in the experiments. The occurrence of a scattering in the polarization-rotated channel is discussed in connection with these results.

Inhomogeneous Magnetic Phase Transition in Fe/Gd Multilayers*

Srajer G.[1], Haskel D.[1], Choi Y.[1], Lee D.R.[1], Lang J.C.[1], Meersschaut J.[2], Jiang J.S.[2] and Bader S.D.[2]

[1] Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA, email: srajerg@aps.anl.gov
[2] Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA

The low-field surface nucleation and evolution of the inhomogeneous magnetic state in strongly coupled Fe/Gd ferromagnetic multilayers is measured via grazing-incidence X-ray magnetic circular dichroism. At $T = 0.7\ T_0$, where $T_0 = 110$ K is the ferromagnetic compensation temperature, the inhomogeneous state nucleates at the surface. At nucleation, the surface state extends tens of interatomic distances into the bulk (~ 200 Å), a direct consequence of the strong interlayer coupling. At $T \sim T_0$, the inhomogeneous state penetrates throughout the bulk, while homogeneous magnetic states occur far below and above T_0. Surface termination has a dramatic effect on the nature of the inhomogeneous state.

*Work at Argonne was supported by the U.S. Department of Energy Office of Science, under Contract No. W-31-109-ENG-38.

Resonant X-ray Scattering of U/Fe Multilayers at XMaS Beamline

Bouchenoire L.[1], Brown S.D.[1], Beesley A.[1], Herring A.[1], Thomas M.[1], Thompson P.[1], Langridge S.[2], Stirling W.G.[3], Mirone A.[3], Lander G.[4], Wilkins S.[4], Ward R.C.C.[5], Wells M.R.[5], Zochowski S.W.[6]

[1] Department of Physics, Oliver Lodge laboratary, University of Liverpool, Oxford street, Liverpool, Merseyside, L69 7ZE, United Kingdom, email: boucheno@esrf.fr
[2] Rutherford Appleton Laboratory, Chilton, Didcot, Oxon OX11 OQX, United Kingdom
[3] European Synchrotron Radiation Facility, BP220, 38043 GRENOBLE Cedex, France
[4] European Institute for Tansuranium Elements, Karlsruhe, Baden-Wurtemberg, Germany
[5] Clarendon Laboratory, University of Oxford, Oxford, Oxon OX1 3PU, United Kingdom
[6] Department of Physics, University College London, London WC1E 6BT, United Kingdom

Magnetic multilayers have been fabricated from many elements. When 3d elements are incorporated in the multilayer stack Giant Magneto Resistance (GMR) is often observed, notably in Fe/Cr, etc. A natural extension of the present multilayer research is to place 5f electrons in these structures. Their wide range of magnetic properties, from localised magnetism (as in UO_2) to itinerant ferromagnetism (as in UFe_2) makes this a rich and potentially important field.

We are now able to produce high quality U/Fe multilayers through DC sputtering. Here, we report on results obtained from a [U31Å/Fe25Å]x30 multilayer through resonant magnetic scattering performed on the XMaS beamline at the ESRF. The asymmetry ratio is defined as $(I^+-I^-) / (I^++I^-)$, where $I^+(I^-)$ is the normalised intensity obtained with circular polarisation and the applied field parallel (anti-parallel) to the beam direction. Asymmetry data will be presented as function of both energy and momentum transfer (**q**). We have identified a uranium moment (~20 % asymmetry ratio at 3.729 keV) with use of resonant magnetic reflectivity at 12 K. Temperature dependence measurements confirm that ~90% of this moment exists at room temperature. Qualitatively, the existing data indicates that the uranium moment is non-uniformly distributed with the layers. The data obtained on tuning the incident energy to a sharp dichroic feature at the Fe K edge also indicates a non-uniform distribution of hybridised Fe. In order to obtain a more quantitive model of the uranium moment distribution it is necessary to acquire acurate values for the absorptive and dispersive parts of the optical constants. To this end, linearly polarised reflectivity has been employed to provide energy and **q** dependent data around the uranium M_{IV} edge and at 7.0 keV. Code has been produced to simultaniously fit to multiple energy data sets using Kramers-kronig transforms of experimental fluorescence data to obtain the dispersive optical constant.

Structural Model for the X-ray Resonant Scattering: Half-Doped Manganites and Magnetite

García J., Subías G., Blasco J., Sánchez M. C. and Proietti M.G.

Instituto de Ciencia de Materiales de Aragón, CSIC-Universidad de Zaragoza, Pza S. Francisco s/n, 50009 Zaragoza, Spain, email: gloria@unizar.es

We review our recent studies on the occurrence of atomic charge localization in magnetite and half-doped manganites. X-ray resonant scattering experiments at the Fe K-edge of magnetite and at the Mn K-edge in $Nd_{0.5}Sr_{0.5}MnO_3$ show the lack of atomic charge localization on the transition-metal atom. The evolution of the intensity of the (002) and (006) forbidden reflections in magnetite across the Verwey transition with the incoming x-ray photon energy, the dependence on the azimuthal angle and the polarization of the scattering process show that the atomic anomalous scattering factors for the octahedral Fe ions are identical to each other, i.e. all of them show the same electronic state [1,2]. On the other hand, the dependence of the x-ray resonant scattering intensity of the satellite reflections (3 0 0), (0 3 0) and the forbidden reflection (0 5/2 0) on the photon energy, the azimuthal angle and the x-ray polarisation in the low temperature phase of $Nd_{0.5}Sr_{0.5}MnO_3$ has been also analysed using a semi-empirical structural model. Contrary to previous claims of atomic charge and orbital ordering (COO), we show that the dipole resonant superlattice reflections can be explained by the presence of two types of Mn sites with different local structure. One of the Mn sites is surrounded by a tetragonal-distorted octahedron of oxygen atoms whereas the other site has a symmetric octahedral environment. The model establishes the lack of integer ionic ordering and the observed resonances are mainly explained as arising from the geometrical local anisotropy. Intermediate valences according to a fractional charge segregation $Mn^{3.42}$-$Mn^{3.58}$ were deduced [3]. Both, the Verwey transition in magnetite and the COO transition in manganites, can be well described as structural phase transitions where a phonon mode condenses giving rise to a new periodicity mainly associated to the oxygen atoms.

References

[1] - J. García, G. Subías, M.G. Proietti, H. Renevier, Y. Joly, J.L. Hodeau and J. Blasco, Phys. Rev. Lett. **85**, 578, (2000)

[2] - J. García, G. Subías, M.G. Proietti, J. Blasco, H. Renevier, J.L. Hodeau and Y. Joly, Phys. Rev. B **63**, 054110, (2001)

[3] - J. Herrero-Martín, J. García, G. Subías, J. Blasco and M.C. Sánchez, submitted to Phys. Rev. B.

A Case for Np 5f Octupole and Hexadecapole Motifs in NpO_2

Lovesey S. W.

Diamond Light Source and ISIS Facility, Rutherford Appleton Laboratory, Oxfordshire OX11 0QX, UK., email: S.W.Lovesey@rl.ac.uk

We consider resonant Bragg diffraction data gathered on NpO_2 held at 12 K and the primary energy tuned to the Np M_4 absorption edge. The actual data used in the interpretation includes both the azimuthal-angle scan observed at the (003) reflection with polarization analysis, and the energy profile around the M_4 edge. The data are consistent with a magnetic order in which the Np dipole moment is zero. In addition, a null value of the Np quadrupole is derived. In the proposed interpretation, diffraction is ascribed to Np 5f octupole and hexadecapole moments.

Atomic Displacement Effects in Resonant 'Forbidden' Reflections

Dmitrienko V.E., Ovchinnikova E.N.[1], Ishida K.[2], Kokubun J.[2], Kirfel A.[3], Collins S.P.[4], Laundy D.[4], Oreshko A.P.[1]

Inst. of Crystallography, 59 Leninskii pr., 119333 Moscow, Russia, email: dmitrien@ns.crys.ras.ru
[1]Moscow State University, Physical Department, 119992,Moscow, Russia
[2]Tokyo University of Science, Noda, Chiba 278-8510, Japan
[3]Mineralogisch-Petrologisches Institut der Universitat Bonn, D-53115 Bonn, Germany
[4]Diamond Light Source, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 OQX, UK
[5]Daresbury Laboratory, Warrington,WA4 4AD, UK

In view of the very different x-ray and phonon energies it is rather surprising that phonons can drastically affect both the intensity and the spectrum of resonant scattering in crystals [1-5]. The reason is that resonant scattering is extremely sensitive, even to very small atomic displacements. Moreover, atomic displacements can change the symmetry of an atomic site, and thus induce an additional anisotropy of the atomic scattering factor near an absorption edge and therefore give rise to extra Bragg reflections, otherwise forbidden.

In this report, we present a survey of vibration effects on the anisotropy of x-ray resonant scattering and discuss thermal-motion-induced (TMI) and point-defect-induced (PDI) 'forbidden' reflections. To this purpose, tensor structure factors and unusual polarization properties of both types of reflections are calculated. Owing to their resonant character, PDI reflections allow for separate studies of both impurity and host atoms of different types. The considered phenomena can provide a very sensitive tool to assess point defects because only those atoms produce contributions to the PDI reflections that are 'distorted' by defects.

Strongly temperature dependent TMI reflections were recently observed in Ge [3,4] and in ZnO [5]. Owing to interference with the temperature-independent contribution, their intensities can increase and decrease with temperature. Drastic changes of the diffraction spectra were found in ZnO, contrary to the rather small changes observed for Ge. Using two different theoretical approaches [6,7] we present some simulations of the temperature dependence and of diffraction spectra for Ge and ZnO.

The work was partly supported by INTAS grant 02-0822.

References

[1] - V. E. Dmitrienko, E. N. Ovchinnikova and K. Ishida, JETP Lett. 69, 938, (1999)
[2] - V. E. Dmitrienko and E. N. Ovchinnikova, Acta Cryst. A 56, 340, (2000)
[3] - J. Kokubun, M. Kanazawa, K. Ishida and V. E. Dmitrienko, Phys. Rev. B 64, 073203, (2001)
[4] - A. Kirfel, J. Grybos and V. E. Dmitrienko, Phys. Rev. B 66, 165202, (2002)
[5] - S. P. Collins, D. Laundy, V. E. Dmitrienko, D. Mannix and P. Thompson, Phys. Rev. B 68, 064110, (2003)
[6] - Y. Joly, Phys. Rev. B 63, 125120, (2001)
[7] - M. Taillefumier, D. Cabaret, A.-M. Flank, and F. Mauri, Phys. Rev. B 66, 195107, (2002)

Ab-Initio Theory of Resonant X-Ray Scattering

Strange P[1], Horne M[1], Arola E[1,2], Winter H[3], Szotek Z[4] and Temmerman W M[4]

[1]School of Chemistry and Physics, Keele University, Keele Staffordshire ST5 5BG, UK,
email: P.Strange@phys.keele.ac.uk
[2]Optoelectronics Research Center, Tampere University, Finland
[3]INFP, Forschungzentrum Karlsruhe,GmbH, Postfach 3640, D-76021, Karlsruhe, Germany
[4]Daresbury Laboratory, CCLRC, Daresbury, Warrington, WA4 4AD, UK

We present a first principles formalism for resonant scattering of circularly polarised x-rays from solids using a minimal amount of mathematics. The theory is split into two parts. Firstly, a good description of the electronic structure of the underlying material is necessary. In our theory this is provided by self-interaction corrected density functional theory and implemented using the fully relativistic LMTO method. Secondly, the theory of the spectroscopy is implemented using standard time dependent perturbation theory to second order in the electron-photon interaction vertex. Using this method *f*-electrons can be described as either localized or delocalized allowing us to study the effect of electron localization and valency of rare earths and their ordered compounds in the x-ray diffraction spectra. Furthermore, being fully relativistic spin-orbit coupling, spin-polarisation and crystal field effects are all treated on an equal footing. Furthermore, in the examples we have looked at all three of Hund's rules are satisfied in the ground state of the material.

The theory will be illustrated with applications to several example materials. These examples will be used to demonstrate the properties of materials that can be deduced from the resonant x-ray spectra. Finally direct comparison with some recent experimental results will be made.

Resonant Magnetic X-Ray Scattering from Transition-Metal Compounds and Actinide Compounds

Igarashi J., Usuda M., Takahashi M.*

Synchrotron Radiation Research Center, Japan Atomic Energy Research Institute, Hyogo 679-5148, Japan, email: jigarash@spring8.or.jp
*Faculty of Engineering, Gunma University, Kiryu, Gunma 376-8515, Japan

The *4p* states of transition metals are delocalized in solids. In usual situations, they attract only minor interest because they are not the states of constituting magnetic or orbital orders. Recently, understanding *4p* states becomes important, since x-ray spectroscopy using *K* edges of transition metals has become popular, in which core electrons are excited to 4p states through the dipole transition. Delocalized nature of *4p* states leads to sensitivilty to electronic structures at neighboring sites. This would make it difficult to analyze the spectra using a simple model. On the other hand, the band structure calculation is expected to work well for describing *4p* states, because electron correlations are small.

In this context, we report ab initio calculations of *K* edge magnetic resonant x-ray scattering (MRXS) spectra in transition-metal and actinide compounds, by taking account of the spin-orbit interaction (SOI) in the LDA scheme. First, we show the result of the MRXS spectra in $KCuF_3$[1] and NiO[2]. These studies demonstrate that the orbital polarization in the *4p* states is induced by their spin polarization through the SOI. Next, we present the calculation of the MRXS spectra in UGa_3, demonstrating that the orbital polarization in the Ga *4p* states is induced by the *5f* orbital moment at neighboring uranium sites through the *p-f* mixing. The study naturally explains the large MRXS intensity found in the experiment[3].

References

[1] - M. Takahashi, M. Usuda and J. Igarashi, Phys. Rev. B **67**, 064425 (2003)
[2] - M. Usuda, M. Takahashi and J. Igarashi, Phys. Rev. B **69**, 014408 (2004)
[3] - D. Mannix, A. Stunault, N. Bernhoeft, L. Paolasini, G. H. Lander, C. Vettier, F. de Bergevin, D. Kacrozowski and A. Czopnik, Phys. Rev. Lett. **86**, 4128 (2002)

Resonant X-ray Scattering as a Tool to Detect Parity-Odd Magnetic Multipoles in Globally Centro-Symmetric Crystals

Di Matteo S.

Laboratorio Nazionale di Frascati, Istituto Nazionale di Fisica Nucleare, Casella postale 13, I-00044 Frascati, Italy, email: Sergio.DiMatteo@lnf.infn.it

A brief outlook is given of the physical quantities detectable by means of resonant x-ray scattering, focusing on the ordinary multipole expansion of the (static) electric and magnetic fields. On this basis I propose a way to identify the parity-odd multipoles, related to the vector potential part of such an expansion, in many centro-symmetric classes of materials with a local inversion-breaking. I put forward two specific examples: Li_2VOSiO_4 and V_2O_3. The first system fulfills all the requirements to individuate magnetic, parity-odd multipoles (i.e., magnetic quadrupoles, toroidal moments, etc.). The second allows the identification of a time-reversal even, parity-odd physical quantity, related to the toroidal moment, through a subtle interference effect.

Notice that such observations would not be possible without the use of a diffraction technique, where the local transition amplitudes are added with a phase factor that can compensate the vanishing 'bulk' effect due to the global symmetry.

The Unresolved Origin of <kkk> Reflections in UAs0.8Se0.2

Bernhoeft N.

CEA Grenoble, DRFMC/SPSMS, 17, rue des Martyrs, F-38054 Grenoble Cedex 9, France,
email: nick_bernhoeft@yahoo.com

The presence of multiple order parameters and their phase coherence on the microscopic scale is a subject of much current debate. In the following it is suggested that the observation of coherent Bragg peaks in multi-**k** magnetic structures, together with detailed studies of their polarisation, energy, azimuth and temperature dependencies, may provide a useful experimental forum to examine some aspects of this problem. Using resonant x-ray scattering, to perform diffraction experiments tuned to the uranium M_4 edge, novel reflections of the generic form <k k k> have been observed in antiferromagnetic $UAs_{0.8}Se_{0.2}$ where **k** = <k 0 0 >, with k = 1/2 reciprocal lattice units. These new reflections, with 10^{-4} of the <k 0 0> magnetic intensities, cannot be explained on the basis of the primary order parameter within standard scattering theory.

It is suggested that the resonant x-ray probe is able to observe the new <k k k> periodicity, arising from the phase coherent superposition of 3 primitive (magnetic) order parameters, through the electric dipole scattering operator. This rationalises the details of measured polarisation, energy and unusual azimuth dependence of the <k k k> peaks in addition to their thermal evolution.

Ground States and Excitations in Half-Doped Manganites: Hard and Soft X-ray Scattering Studies

Hill, J.P.[1]

[1] Department of Physics, Brookhaven National Laboratory, Upton, NY 11973, USA,
email: hill@bnl.gov

A complete understanding of the electronic properties of a given system requires an understanding of both the ground states and the excitations of that system. Here, we apply resonant x-ray scattering techniques to both aspects of this problem, focusing on the half-doped manganites. K-edge elastic scattering studies were performed on $Pr_{0.6}Ca_{0.4}MnO_3$ [1]. This system has been previously argued to exhibit the checkerboard charge and orbitally ordered phase with a CE-type magnetic structure. K-edge studies are sensitive to the environment of the Mn ion and a detailed analysis of the resonant scattering reveals a partial d-occupancy on all Mn sites with $3x^2-r^2/3y^2-r^2$ type orbital order on the 'Mn^{3+}' sites and x^2-y^2 occupancy on the 'Mn^{4+}' sites. No evidence for a 1s core level shift was observed, suggesting that the charge disproportionation is small. To investigate this further, studies were also performed at the Mn L-edge [2]. Here, resonant scattering probes the 3d orbitals directly. Large enhancements were observed at the magnetic and orbital wavevectors. The magnetic correlation length was found to be significantly larger than the orbital correlation length. In addition, a large (3eV) shift in the spectral weight was observed between the magnetic and orbital resonant lineshapes. A relaxed charge order model is proposed to explain these results. Finally, inelastic x-ray scattering experiments were performed at the Mn K-edge on this and related manganites [3]. Significant changes were observed as a function of temperature, up to energies of several eV – well above thermal energies, demonstrating the extreme sensitivity of the electronic structure to weak perturbations in these materials. The changes are correlated with the magnetic transitions.

Work performed at Brookhaven was supported by the US DOE, Division of Materials Science, under contract no. DE-AC02-98CH10886. The author particularly wants to acknowledge his collaborators, S. Grenier and K.J. Thomas, for their participation in this work.

References

[1] - S. Grenier, et al. cond-mat/0305216
[2] - K.J. Thomas et al., cond-mat/0311553
[3] - S. Grenier et al., unpublished work

Imaging Spiral Magnetic Domains in Rare Earth Metals*

Lang J.C.[1], McWhan D.[2], Lee D.R.[1], Haskel D.[1] and Srajer G.[2]

[1]Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA, email: lang@aps.anl.gov
[2]NSLS, Brookhaven National Lab (retired), Upton, NY, 11973, USA

This work presents images showing the temperature dependence of the spiral magnetic domains in Dy and Ho metals. In these materials, the moments order ferromagnetically in the hexagonal basal plane, with the magnetization direction rotating between successive atomic layers forming a helix. The sense of this rotation can be either right or left handed leading to the formation of chirality domains. At magnetic Bragg diffraction peaks, circularly polarized X-rays are sensitive to the handedness of such a helix (i.e. either right or left handed). The spiral domain structure can therefore be imaged by combining phase retarding optics with micro-diffraction techniques. This contrast between the chiral domains, occurs both for resonant and non-resonant magnetic scatting. Near the $L_{2,3}$ absorption edge resonances, however, the differences in the scattering intensity vary dramatically. On either side of the enhancement, the contrast is strongly suppressed to ~-10%, while at the enhancement peak and far from the edge the contrast approaches ~85%. Images showing the energy, temperature, and wave vector dependence of the domain structure will be presented.

*Work at Argonne was supported by the U.S. Department of Energy Office of Science, under Contract No. W-31-109-ENG-38.

Soft X-ray Resonant Diffraction from Single Crystals

Hatton Peter D.

Department of Physics, University of Durham, UK, email: p.d.hatton@Durham.ac.uk

Resonant X-ray diffraction has developed into a major technique applicable to many materials. We will report on the extension of resonant diffraction into the soft X-ray region (500 –1500 eV). We have published examples of the huge resonant enhancements of charge and magnetic scattering that can be obtained at the *L*-edges of 3*d* transition metal oxides [1,2]. In this talk we will review the first resonant soft X-ray diffraction observations of charge, spin and orbital ordering from bulk single crystals. We have recently studied the low temperature phase of $La_{0.5}Sr_{1.5}MnO_4$ that displays charge, spin and orbital ordering. Energy scans at constant wavevector have been compared to theoretical predictions and show that at all temperatures there are two separate contributions to the observed scattering, direct Goodenough orbital ordering and strong cooperative Jahn-Teller distortions of the Mn^{3+} ions. Our results also suggest a strong coupling between the spin and orbital degrees of freedom. We have also applied this technique to the study of other materials such as holmium and $UAs_{1-x}Se_x$ (x = 0.15) where incommensurate magnetic diffraction has been observed at the Ho M_V and U N_{IV} and N_V edges.

References

[1] - S.B. Wilkins, P.D. Hatton, M.D. Roper, D. Prabhakaran and A.T. Boothroyd. Phys. Rev. Lett. **90** (18) 187201 (2003)

[2] - S.B. Wilkins, P.D.Spencer, P.D. Hatton, S.P. Collins, M.D. Roper, D.Prabhakaran and A.T. Boothroyd Physical Review Letters. **91** (16) 167205 (2003)

Synchrotron X-ray Diffraction Measurements in High Magnetic Fields and Low Temperatures

Katsumata K.

SPring-8/RIKEN, Mikazuki, Sayo, Hyogo 679-5148, Japan,
e-mail : katumata@postman.riken.go.jp

With the use of high intensity X-rays from the SPring-8, X-ray diffraction by magnetic materials are now routinely measured at the beamline BL19LXU. It is then natural to extend the measurement to higher magnetic fields and to lower temperatures. We have constructed an X-ray diffractometer equipped with a 15 T superconducting magnet in conjunction with a dilution refrigerator. In this presentation, I will report on our recent results of X-ray magnetic and charge scattering measurements on magnetic materials obtained in high magnetic fields and/or at very low temperatures.

1) Observation of the spin-flop transition in MnF_2 by X-rays

It is well established that the intensity of neutrons diffracted by a magnetic material depends on the angle the magnetic moment in the sample makes with the scattering vector. We have studied the dependence of X-ray magnetic diffraction intensity on the scattering vector. For this purpose, we measured the spin-flop transition in a uniaxial antiferromagnet MnF_2. We found that the magnetic (3, 0, 0) Bragg intensity decreased steeply at about 9.3 T at which the spin-flop transition has been observed from bulk measurements.

2) X-ray diffraction measurements in the high field phase of a spin-Peierls compound $CuGeO_3$

We have measured the temperature dependence of the lattice incommensurability, δ in the high field phase of $CuGeO_3$ at fixed fields. At 12.6 T, slightly above the critical field from the commensurate to incommensurate phases, we observed that δ became small with increasing temperature, in accordance with the observation reported before. In contrast, at 15 T, δ is almost independent of temperature. This observation shows that strong magnetic fields suppress the thermal fluctuation of the incommensurate state.

3) Observation of lattice instability at the field induced phase transition of the spin gapped compound $Cu_2(C_5H_{12}N_2)_2Cl_4$

The ground state of the title compound is a singlet and the material shows a field induced magnetic ordering at low temperatures below about 0.8 K. We have measured the field dependence of the lattice constants in this compound at about 40 mK and found the lattice constant showed a jump at about 7.3 T. This observation is consistent with a theoretical prediction of the lattice instability associated with the field induced phase transition in a quantum antiferromagnet.

The results reported here have been obtained in collaboration with C. Berthier, T. Bizen, S. Goto, M. Hagiwara, T. Hara, I. Harada, T. Ishikawa, S. Kimura, H. Kitamura, J. E. Lorenzo, S. W. Lovesey, M. Matsuda, T. Matsushita, H. Mayaffre, H.-J. Mikeska, T. Nakamura, Y. Narumi, Y. Nishiyama, H. Ohashi, T. Ohata, S. Shimomura, Y. Tabata, K. Takeshita, K. Tamasaku, T. Tanaka, Y. Tanaka, M. Yabashi and I. Yamada.

High Pressure Resonant Magnetic X-ray Scattering: Dream or Reality?

Braithwaite Daniel

CEA Grenoble, Departement de Recherche Fondamentale sur la Matière Condensée, SPSMS, 17 rue des Martyrs, 38054 Grenoble Cedex 9, France, email: braithwaite@drfmc.ceng.cea.fr

Synchrotron radiation experiments and high pressure are two well suited techniques due to the fact that the synchrotron beam can be focussed on a small sample area, typically 100x100 microns, compatible with sample sizes used in high pressure, including with the diamond anvil cell. The scope for magnetic diffraction experiments under pressure is enormous, for the study of many magnetic phenomena beyond the pressure range achievable with neutron scattering techniques (usually 2-3 GPa). However several specificities of magnetic X-ray diffraction, notably the low energies, the large scattering angles, and the weak intensity of the magnetic signal, make this technique more difficult than most, to implement under pressure. We describe a specially designed pressure set-up to take into account these aspects and show initial results of resonant magnetic scattering under pressure on the $Ce(Fe_{1-x}Co_x)_2$ system. We also demonstrate the usefulness of the set-up which allows the pressure to be changed and measured at low temperature for other measurements such as structural studies and resonant x-ray absorption studies. This has been used for a recent study on samarium sulfide, allowing the determination of the lattice parameter and samarium valence changes at low temperature (T=4K) in the insulating, metallic and magnetic phases.