**PROGRESS REPORT
PERIOD 9/2003-3/2004
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Title: Polarized x-rays as a probe of spin polarization and interface and confinement effects: Theoretical approach

Grant Number DE-FG02-03ER460097

1. Personnel

Due to the somewhat late confirmation of the grant, we had a slight delay in starting the search for a Research Associate (NIU does not allow posting of advertisements prior to receiving the grant). The advertisement was placed in Physics Today and distributed electronically. The following shortlist of candidates has been selected:

Gonzalo Alvarez

Jun Li

Serkan Erdin

After confirmation of affirmative action a candidate will be selected.

2. Results obtained in preceding period.

I. Spin polarization in CrO₂

Spin polarization in half-metallic CrO₂

Michel van Veenendaal and A. J. Fedro, submitted to Microscopy and Microanalysis (2/11/2004)

Spin Polarization in CrO₂: Competition between quasiparticle and local-moment behavior

Michel van Veenendaal and A. J. Fedro, submitted to Phys. Rev. Lett. (12/3/03, manuscript LM8973)

In materials science, people are investigating materials with a high degree of spin polarization that could form the basis for a future device technology where the spin adds a degree of freedom with respect to the charge-based silicon technology [1]. One material that is being investigated is CrO₂, well known from its use in magnetic tapes. According

to several LDA-based electronic structure calculations [2-4], CrO₂ is expected to have a spin polarization close to 100% several eV around the Fermi level.

Polarized and spin-resolved spectroscopic techniques are crucial in obtaining detailed information on the spin polarization. Below the Fermi level, spin-resolved photoemission [5] shows indeed a spin polarization close to 100%, see Fig. 1. However, above the Fermi level, recent spin-resolved circularly-polarized resonant photoemission experiments [6] show a behavior different from that expected from the LDA-based calculations. Within 0.2 eV above the Fermi level, the spin polarization is close to 100%. However, at higher energies above the Fermi level, the spin polarization rapidly decreases to around 50%. The value of 50% is close to what one would expect of electron addition to a local $S=1$ moment. CrO₂ therefore show a strong competition between quasiparticle and local moment character.

We have performed model calculations that take into account the dual character of the electronic structure of CrO₂. The results are given in Fig. 1. Qualitatively the results can be interpreted as follows. An added spin-up electron can form a quasiparticle with the positively polarized spin background. This leads to a quasiparticle peak at the Fermi level. However, an added spin-down electron is unable to form a quasiparticle with the Fermi sea of spin-up electrons. Therefore, close to the Fermi level only 100% spin-polarized quasiparticle weight is found. However, at higher energies above the Fermi level, the local moment character starts to dominate. To a local moment of $S=1$, one can add an spin-up electron forming a $S=3/2$, $S_z=3/2$ local moment. In the case of a spin down electron one can form $S=3/2$ and $1/2$ local moments, both with $S_z=1/2$. The $S=3/2$ moment is close to the Fermi level, leading to a spin polarization of 50%. This is a many-body Slater determinant effect not properly taken into account in the single Slater-determinant based LDA approaches.

In conclusion, we have shown the importance of including the competition between local moment and quasiparticle character in describing the electronic structure of CrO₂.

- [1] For a review see S. A. Wolff, *et al.*, Science **294**, 1488 (2001).
- [2] I. Mazin, D. J. Singh, and C. Ambrosch-Draxl, Phys. Rev. B **59**, 411 (1999).
- [3] M. Korotin, *et al.*, Phys. Rev. Lett. **80**, 4305 (1998).
- [4] L. Craco, M. S. Laad, and E. Müller-Hartmann, Phys. Rev. Lett. **90**, 237203 (2003).
- [5] K. P. Kämper, *et al.*, Phys. Rev. Lett. **59**, 2788 (1987).
- [6] D. J. Huang *et al.*, Phys. Rev. B **67**, 214419 (2003).

FIG. 1. The lower panel shows the spin-polarized density of states including the competition between quasiparticle and local-moment behavior as described in the text. The upper panel shows the spin polarization (blue). For comparison, the experimental spin polarization from spin-resolved photoemission [5] (triangles) and spin-resolved polarized resonant O 1s Auger spectroscopy [6] (diamonds), and the theoretical spin polarization obtained with the LDA plus dynamical mean field [4] (green) are shown.

Soft resonant x-ray scattering study of magnetic and orbital correlations in a manganite near half-doping

K. J. Thomas, J. P. Hill, Y.-J. Kim, S. Grenier, P. Abbamonte, L. Venema, A. Rusydi, Y. Tomioka, Y. Tokura, D. McMorrow, G. A. Sawatzky, and M. van Veenendaal, submitted to Phys. Rev. Lett.

Recently, it has been demonstrated that resonant diffraction at the Mn *L*-edges may be utilized to study magnetic and orbital order in the manganites. The Mn *L*-edge resonances involve strong dipole transitions from the 2*p* core levels to unoccupied states within the 3*d* band. The resonant enhancement of magnetic scattering at the Mn *L*-edge is significantly greater than at the *K*-edge. Furthermore, the structure of the resonant diffraction line shape provides a direct spectroscopy of the Mn 3*d* band in the presence of correlations. In this letter, we present a resonant x-ray diffraction study of a near half-doped manganite at the Mn *L*-edge which permits the first direct comparison between magnetic and orbital correlations. Our data provide evidence against a charge ordering into distinct Mn³⁺ and Mn⁴⁺ sub-lattices in Pr_{0.6}Ca_{0.4}MnO₃, and indicate a more complex ground state. First, we find that the magnetic correlations are significantly longer-ranged

than the orbital correlations. Second, the spectral line shapes and the difference in intensity between magnetic and orbital scattering disagree with the ionic CE picture. Rather, we suggest that while the e_g electron is still localized, it is shared between neighboring lattice sites[1].

This work was done in collaboration with John Hill's x-ray scattering group at Brookhaven National Laboratory.

[1] M. van Veenendaal and A. J. Fedro, Phys. Rev. B **59**, 15 056 (1999).

FIG 2. Comparison between the measured and calculated resonance line shapes in $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$. The upper half shows the magnetic scattering and the lower half shows the orbital scattering.

III. Resonant Inelastic X-ray Scattering

Theory of indirect resonant inelastic x-ray scattering

J. van den Brink and M. van Veenendaal, submitted to Phys. Rev. Lett. (11/19/03, manuscript LL9336).

Resonant Inelastic X-ray Scattering (RIXS) is a technique that has lately come to the forefront. Being a photon in-photon out technique, RIXS is much less surface sensitive than, e.g. photoelectron spectroscopy. Although the technique is a core level spectroscopy with the advantage to chemical specificity, the final state contains no core hole and can provide direct information on valence shell excitations. In the last few years experiments have been performed at the K -edge in transition-metal compounds, such as the high- T_c compounds and the manganites. In these systems, valence shell excitations are created by the interactions of the $1s$ core hole and the $3d$ valence electrons. In a collaboration with Jeroen van den Brink from the University of Leiden, the Netherlands, we have shown that under certain conditions the RIXS cross section can be related to the dielectric function.

3. Summary of future research

This proposal investigates the use of (polarized) x-rays in the study of spin polarization and interface and confinement effects. Progress has been made in the understanding of the electronic structure important for spin-based electronics, such as CrO_2 and the manganites.

I. Resonant Inelastic X-ray Scattering (RIXS)

In the coming year, a significant amount of time will be devoted to further establishing the theory of RIXS. One can distinguish two different kinds of RIXS. First, direct transition can be made into strongly correlated bands, e.g. at the L -edges in transition metals and their compounds. Here the scattering is mainly determined by the combination of dipole operators for absorption and deexcitation. This process is not well understood, as evidenced by the recent discussion on the RIXS data on NaV_2O_5 .

On the other hand, one can make transitions into wide bands. Final state excitations are then created due to the interaction between the core hole and the valence shell. Recent data on several manganites by Grenier *et al.*[3] show a dependence of the integrated inelastic spectral weight on the magnetic and charge ordering. The ordering appears to be of more importance than the metallic or insulating properties. Although we have some qualitative interpretations, it is important to obtain a more detailed understanding of the RIXS process. Although some efforts have been made to relate the RIXS cross section to the dielectric function, results have only been obtained in certain limits.

Other aspects that need to be investigated are: What is the role of the interaction between the core hole and the excited electron? Generally, these interactions are neglected and only the interaction with the strongly correlated band is taken into account.

What is the effect of interactions with other excitations, such as magnons and phonons on the final-state excitations, and in particular their q -dependence?

[1] G. P. Zhang et al., Phys. Rev. Lett. **88**, 077401 (2002).

[2] M. van Veenendaal and A. J. Fedro, Comment on “Correlation Effects in Resonant Inelastic X-ray Scattering of NaV_2O_5 ”, submitted to Phys. Rev. Lett. (6/2/03, manuscript LFK892)

[3] S. Grenier, J. P. Hill, *et al.*, unpublished.

II. Spin polarization and (polarized) x-ray spectroscopies

The theory of spin polarization described above will be further developed. Spectroscopy shows the importance of including the competition between the local moment and quasiparticle character in the description of the spin polarization. This effect is not properly taken into account in LDA based methods, including LDA+U and LDA plus Dynamical Mean Field. The spin-polarized Density of States (DOS) below the Fermi level can be probed with spin-resolved photoemission. The unoccupied spin-polarized DOS is less easily accessible by spectroscopy. Information has been obtained using spin-resolved circularly polarized resonant photoemission. This technique is still not fully understood in particular for intermediate states where the magnetic shell is not fully occupied. I will further investigate the use of these techniques in the study of the spin polarization. In addition, I will explore the use of resonant inelastic x-ray scattering as a probe of spin polarization and spin-flip processes.

CURRENT FEDERAL GRANTS

Research on the interpretation of X-ray spectroscopy

PI M. van Veenendaal, awarded by the University of Chicago/Argonne National Laboratory. Award amount \$40,541.

Theoretical support for synchrotron-related works.

PI M. van Veenendaal. Project in the Theory Group of the Materials Science Division at Argonne National Laboratory led by Mike Norman. Award amount: \$9,174

GRANTS PENDING

Spin polarization in materials with competing quasiparticle and local moment behavior.

Submitted October 2002 with PI M. van Veenendaal to the Materials Theory section of the Materials Research Division of the National Science Foundation. Requested amount \$370,821.

NER: X-ray magnetic dichroism as a probe of photoinduced magnetism in molecule-based magnets

Submitted October 2002 with PI M. van Veenendaal to NSF-03-043 “Nanoscale Science and Engineering (NSE)” of the National Science Foundation. Requested amount \$84,204.

NIRT: Nanocharacterization of electronic and magnetic behavior with polarized x-rays and e-beams.

Submitted October 2002 with PI M. van Veenendaal and co-PI's Zhili Xiao, D. Brown, O. Chmaissem, and Y. Ito to NSF-03-043 “Nanoscale Science and Engineering (NSE)” of the National Science Foundation. Requested amount \$1,604,532.