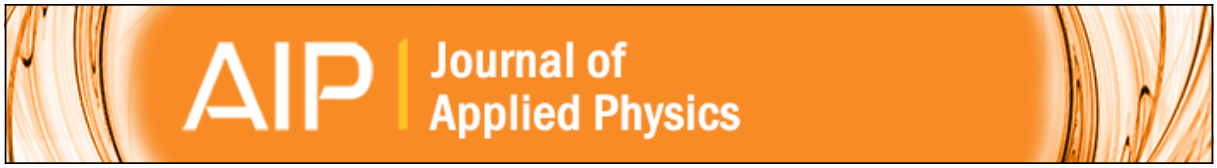

Observation of orbital moment in NiO using magnetic x-ray scattering

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The spin and orbital contribution to the total magnetization have been measured in NiO by exploiting the polarization dependence of nonresonant x-ray magnetic scattering. Although the orbital moment is usually neglected, we observed that the orbital moment actually plays an important role with a contribution of 17% to the total magnetization density. By performing azimuthal scans, it was found that spin and orbital moments are collinear. © *1999 American Institute of Physics.* [S0021-8979(99)59208-6]

I. INTRODUCTION

There has been a renewed interest in transition metal oxides due to the discovery of giant magnetoresistance and recent findings about orbital ordering.[1,2] Furthermore monoxides of $3d$ metals can be used as test systems for band-theory models. The measurement of the orbital moment in these materials can provide an important piece of information on their electronic and magnetic properties.

In this work, we have used the unique properties of magnetic x-ray scattering to measure the orbital moment contribution to the total magnetization density in NiO. Indeed the spin and orbital moment parts to the total magnetic moment contribute differently to the scattering cross-sections.[3] When the incoming beam is polarized perpendicular to the scattering plane, the scattering from orbital moments rotates the polarization while the scattering from spins contributes to both channels, rotated and nonrotated.

NiO is an antiferromagnetic insulator of type II at room temperature.[4] It has a crystal structure of the NaCl type ($a_0 \sim 4.177$ Å). The type-II antiferromagnetic structure is characterized by ferromagnetic $\underline{(111)}$ planes, with magnetic moments aligned along a $\langle 112 \rangle$ direction, stacked antiferromagnetically along the $\langle 111 \rangle$ direction.[5] Due to the cubic symmetry of the system, four equivalent propagation vectors of type $\langle 1/2\ 1/2\ 1/2 \rangle$ exist,[6] which give rise to four magnetic T domains. Within each of these four T domains, three S domains can be found, corresponding to the three equivalent directions of the magnetic moments.

Because of its simple magnetic structure and high T_N value of 523 K, NiO has been chosen as the first test sample for magnetic x-ray scattering.[7]

II. EXPERIMENTAL METHOD

The complete development of the nonresonant x-ray scattering cross-sections can be found in literature.[3,8,9] In contrast to neutron diffraction,[10] the nonresonant magnetic x-ray scattering method relies on the polarization analysis of scattered photon beams to extract unambiguously the L and S contributions to the total magnetic moment. It is therefore important to use high linearly polarized x rays. Such x-ray beams are available from insertion devices of 3rd generation synchrotron sources.

The polarization analysis technique makes use of Bragg diffraction at $2\theta = 90°$ from a crystal analyzer. Thomson charge scattering at $2\theta = 90°$ only scatters light which is perpendicular polarized to the scattering plane of the analyzer crystal. If the scattering plane of the sample is parallel to the one of the crystal analyzer only the polarization component perpendicular (σ) to the scattering plane of the sample is measured. When the two scattering planes are perpendicular the detector measures the intensity of the polarization component which was in the scattering plane of the sample (π).

The possibility to adjust the energy to perform polarization analysis together with the necessity of a highly polarized photon beam and the rather weak scattering amplitude of magnetic x-ray scattering (3 orders of magnitude weaker than charge scattering) make it necessary to work at a synchrotron light source.

The present work was thus performed at the magnetic scattering beamline ID20 at the European Synchrotron Radiation Facility (ESRF). This beamline provides an incident power on the sample at 7.84 keV of approximately 2.0×10^{12} ph/s at 200 mA, with a beamsize of 0.3×0.2 mm^2. In the geometry used in this experiment the incoming photon beam was polarized perpendicular to the scattering plane (σ polarization).

The NiO crystal [with a (111) surface] was mounted on a diffractometer that can be used in four circle geometry together with an azimuthal setup and which also provides polarization analysis of the scattered beam.[11] PG (006) was selected as analyzer crystal, to perform polarization analysis at 7.84 keV.

III. EXPERIMENTAL RESULTS

In this first study, we have concentrated on the specular magnetic reflections (1/2 1/2 1/2), (3/2 3/2 3/2), and (5/2 5/2 5/2). Intensities were collected by rocking the NiO crystal

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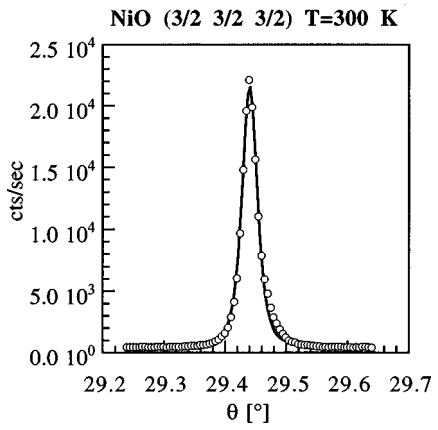

FIG. 1. Rocking curve of the (3/2 3/2 3/2) magnetic reflection.

both with and without polarization analysis. The polarizer crystal was also rocked when measuring the (3/2 3/2 3/2) to check that proper integrations were achieved thanks to the mosaic spread of the PG (006) crystal. Figure 1 shows one of the reflections without polarization analysis. Measuring about 20.000 cts/s in the magnetic peak compared to 2 cts/min measured during the first experiment by de Bergevin and Brunel in 1972, shows the important role of the development of third generation synchrotron light sources in the field of magnetic x-ray scattering.[7]

Comparison of intensities with and without polarization analysis allows a good determination of the polarization analyzers efficiency, which is important to obtain absolute intensities.[12]

The intensities of the (3/2 3/2 3/2) reflection were followed as a function of temperature (Fig. 2). All intensity disappears at $T_N = 520$ K, which demonstrates the magnetic character of the observed intensities.

A. Observation of magnetic *S* domains

The measured reflections all belong to the same magnetic *T* domain, characterized by the propagation vector $\langle 1/2\ 1/2\ 1/2\rangle$. But the measured intensity within one *T* domain is in fact the incoherent sum of the three associated *S* domains.

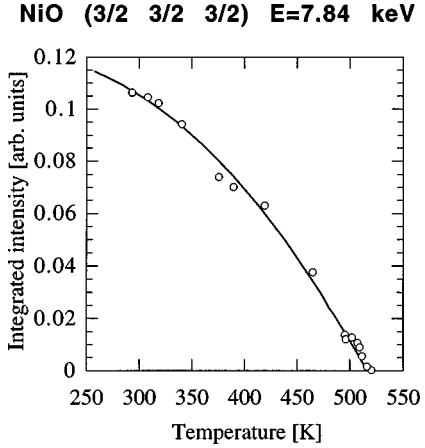

FIG. 2. The integrated intensities of the (3/2 3/2 3/2) magnetic reflection as a function of temperature. Intensity disappears at about 520 K.

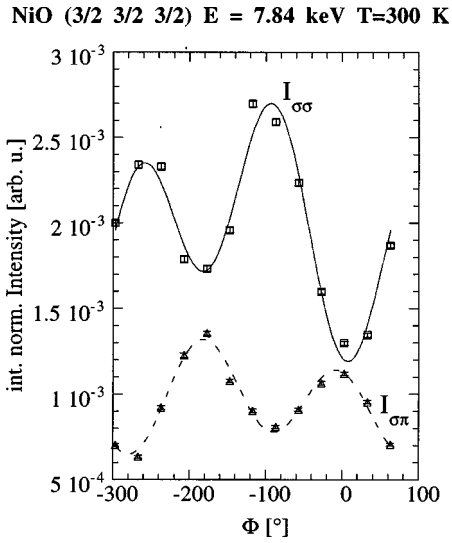

FIG. 3. Normalized integrated intensities of the (3/2 3/2 3/2) reflection as a function of the Φ angle at 7.84 keV. The two polarized components are out of phase by $\pi/2$. The full lines show the calculated modulation of the three *S* domains.

The contribution of each *S* domain to the scattering amplitude[12] can be expressed as:

$$F_{\sigma\sigma} = -\alpha_i r_0 \frac{i\hbar\omega}{mc^2} \sin 2\theta \sin(\Phi) S(\mathbf{Q}),$$

$$F_{\pi\sigma} = -\alpha_i r_0 \frac{i\hbar\omega}{mc^2} \sin 2\theta \sin\theta [\cos(\Phi) S(\mathbf{Q}) + \cos(\mathbf{\Phi} + \mathbf{\Phi}_0) \mathbf{L}(\mathbf{Q})], \tag{1}$$

where r_0 is the classical electron radius, $\hbar\omega$ the photon energy, 2θ is the scattering angle and S_i and L_i with $(i = 1,2,3)$ are the components of $\mathbf{S}(\mathbf{Q})$ the Fourier transform of the spin density, and $\mathbf{L}(\mathbf{Q})$ the Fourier transform of an operator containing the orbital moment. The three basic vectors are given in the publication of Blume.[3] At $Q = 0$, $\mathbf{L}(\mathbf{Q})$ gives directly the orbital moment. The origin of Φ is the spin direction in the scattering plane and Φ_0 is an angular off-set between *S* and *L*. α_i with $i = 1,2,3$ is the relative volume of the domains. In the *S* multidomain sample of NiO we have a superposition of these terms with a phase shift of $2\pi/3$ between the domains. The scattered intensities are thus combinations of $\sin^2(\Phi)$ and $\cos^2(\Phi)$.

By performing azimuthal scans (intensity as a function of Φ) about the surface normal [111], we measured the modulation of the intensities coming from the three *S* domains in the two polarization channels (Fig. 3). The phase shift of $\pi/2$ between the two polarization channels indicates that there is a collinear alignment of the spin and orbital moment $(\Phi_0 = 0)$. The observed intensities not only depend on the spin directions relative to the incoming polarization, but also on the relative volume of the three *S* domains. Best adjustments of the experimental points were obtained for relative domain volumes of 0.26 ± 0.03, 0.23 ± 0.03, and 0.50 ± 0.02 for the sample region probed by the x-ray beam.

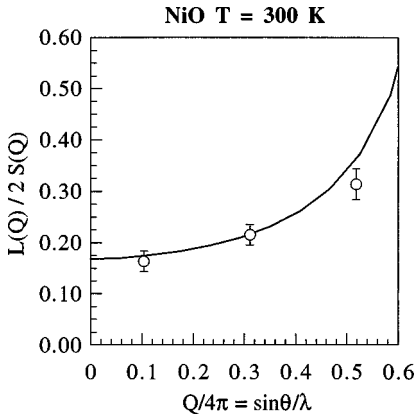

FIG. 4. Variation of $L(Q)/2S(Q)$ as a function of sin θ/λ. The continuous line is established using an estimation of Blume (Ref. 13) and is adjusted to fit the data. At $Q=0$ the $L(Q)/2S(Q)$ value is 0.17.

B. *L* and *S*

To extract the information on the orbital moment and the spin out of the two measured intensities $I_{\sigma\sigma}$ and $I_{\sigma\pi}$, the intensities were averaged over the angle Φ ($\overline{I_{\sigma\sigma}}$ and $\overline{I_{\sigma\pi}}$). These averaged intensities correspond to $\langle \sin^2(\Phi)\rangle = \langle \cos^2(\Phi)\rangle = 1/2$ in the magnetic cross-section [square of Eq. (1)]. Furthermore we have corrected the intensities by the width of the rocking curve of the analyzer crystal ($W_{\sigma\sigma}$ and $W_{\sigma\pi}$). Using Eq. (1) the ratio L/S can be established straightforwardly

$$\frac{L}{S} = \frac{1}{\sin(\theta)} \sqrt{\frac{\overline{I_{\sigma\pi}} W_{\sigma\pi}}{\overline{I_{\sigma\sigma}} W_{\sigma\sigma}}} - 1. \tag{2}$$

Figure 4 shows the ratio $L/2S$, which reflects the magnetic contributions of spin and orbital moment that we obtained experimentally for the three reflections. The solid line is established using Blumes[13] form factor estimations. The extrapolation to $Q=0$ gives an $L/2S$ ratio of 0.17. This shows clearly that the orbital moment plays a role in the magnetization of NiO. The increase of $L/2S$ at large Q shows the broader spatial extent of the spin density.

By comparing the intensities of the magnetic reflections with the intensities of the charge peaks we were able to put the magnetic structure factors on an absolute scale.[12] From the extrapolated values at zero momentum transfer we obtained a total magnetic moment of $\langle \mu \rangle = \langle L(0)\rangle + 2\langle S(0)\rangle = 2.2 \pm 0.2 \mu_B$ in good agreement with neutron data.[10]

IV. CONCLUSION

By using magnetic x-ray scattering, we have been able to measure the orbital momentum contribution to the total magnetic density. The L/S ratio of 0.34 at $Q=0$ shows that the orbital moment in NiO is far from being quenched and that more studies are needed to understand its origin. The large orbital moment value should be taken into account by band model calculations in NiO.

A second interesting result of this study is the fact that spin and orbital moments are collinear which is expected from spin-orbit coupling of a free $3d^8$ atom.

Further work is needed in other oxides, especially CoO, where orbital momentum is expected to be larger.

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[1] Y. Muraki *et al.*, Phys. Rev. Lett. **81**, 582 (1998).
[2] Y. Muraki *et al.*, Phys. Rev. Lett. **80**, 1932 (1998).
[3] M. Blume and D. Gibbs, Phys. Rev. B **37**, 1779 (1988).
[4] C. G.Shull, W. A. Strauser, and F. O. Wollan, Phys. Rev. **83**, 333 (1951).
[5] J. Baruchel, M. Schlenker, K. Kuusowa, and S. Saito, Philos. Mag. B **43**, 853 (1981).
[6] W. Roth, J. Appl. Phys. **31**, 2000 (1960).
[7] F. D. Bergevin and M. Brunel, Phys. Lett. **39A2**, 141 (1972).
[8] M. Brunel and F. D. Bergevin, Acta Crystallogr. A **37**, 324 (1981).
[9] M. Blume, J. Appl. Phys. **57**, 3615 (1985).
[10] H. A. Alperin, J. Phys. Soc. Jpn. **17B-III**, 12 (1962).
[11] A. Stunault *et al.*, J. Synchrotron Radiat. **5**, 1010 (1998).
[12] V. Fernandez *et al.*, Phys. Rev. B **57**, 7870 (1998).
[13] M. Blume, Phys. Rev. **124**, 96 (1961).