Simulations of magnetic Bragg scattering in transmission electron microscopy

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We have modeled the magnetic Bragg scattering in two antiferromagnetic materials, NiO and LaMnAsO, using transmission electron microscopy. Experimentally, Loudon detected these weak magnetic phenomena in NiO. As a more difficult situation with a lower concentration of magnetic elements and higher concentration of heavier non-magnetic elements that significantly scatter, we did computations for the LaMnAsO compound in order to compare our computational replication of Loudon’s experimental data. Additionally, we have discussed the antiferromagnetic Bragg spot’s thickness and voltage dependency for both compounds. We used two computational methods, one assuming a static lattice with smeared Debye-Waller potentials and the other explicitly taking into account the atomic vibrations within the quantum excitations of phonons model (thermal diffuse scattering). According to the structural study, the antiferromagnetic Bragg spot in NiO is located between the (111) and (000) reflections. However, in LaMnAsO, it is located at the site of the (110) reflection in the diffraction pattern, which is a forbidden reflection in the crystal structure. According to calculations, the magnetic Bragg spot in NiO has an intensity that is much greater than thermal diffuse scattering at room temperature. The magnetic Bragg spot for LaMnAsO is weaker than the thermal diffuse scattering at room temperature, but its identification can be made easier at lower temperatures.

Index Terms—Antiferromagnetic materials, Transmission Electron Microscopy, Magnetism, Bragg scattering.

Rapid advancements in magnetic nanoengineering need the use of characterization techniques with high spatial resolutions that can describe magnetic phenomena at the atomic level. One of the options for such high-resolution material characterization is transmission electron microscopy (TEM). Due to the fact that an electron beam is made up of moving charged particles, it can be compared to an electrical current that is affected by the magnetic and electric fields of the sample. It can be utilized to image the magnetic structure of materials by using this interaction. Ehrenfest’s theorem serves as the theoretical foundation for this way of visualizing the magnetic structure [1], [2].

Antiferromagnetic materials (AFM) with collinear magnetic moments in particular have attracted attention recently due to the downsizing of magnetic technology and the possible spintronic applications they may have [3]. It was demonstrated in Loudon’s study [4] that NiO thin films allow for the TEM observation of an antiferromagnetic Bragg spot. Despite being 10000 times less powerful than Bragg peaks formed from the structure, the antiferromagnetic reflection of magnetic Bragg scattering was discovered to stand out sharply from the thermal diffuse scattering (TDS) background. Loudon additionally calculated the oscillation of the magnetic reflection caused by dynamical diffraction to be approximately 236 nm using kinematic approximation. These findings demand additional investigations utilizing a more complex computational model to comprehend the relationship between antiferromagnetic Bragg spot visibility of the thickness, accelerational voltage, and non-zero temperature.

In this study, we investigate the dependence of the intensity of antiferromagnetic Bragg spots in LaMnAsO (space group $P4/mnm$) and NiO (space group $C_{2/c}$) on sample thickness and acceleration voltage using a multislice simulation framework based on the paraxial Pauli equation [5] [6]. These simulations were also carried out with the explicit inclusion of atomic vibrations, resulting in TDS, for a chosen sample thickness of approximately 123 nm and an acceleration voltage equal to 300 kV, in order to compare the intensity of magnetic Bragg spots to the intensity of the TDS background.

Edström et al. [5] [6] provided a detailed description of the Pauli multislice methodology that was employed in this work, and is founded on an approximate paraxial solution of Pauli’s equation,

$$\frac{\partial \Psi}{\partial z} = \frac{im}{\hbar} (\hbar k + eA_z)^{-1} \left( \frac{\hbar^2 \nabla^2_{xy}}{2m} + \frac{ie\hbar}{m} A_{xy} \cdot \nabla_{xy} \right. $$

$$- \frac{\hbar keA_z}{m} - \frac{eV}{2m} \sigma \cdot \mathbf{B} + eV \left. \right) \Psi, \quad (1)$$

as an alternative to Schrödinger’s equation in the conventional multislice approach [7]. The electrostatic potential $V$, the in-plane gradient operator $\nabla_{xy}$, the Pauli spinor $\Psi$, which is a wave function with both spin-up and spin-down components, and the relativistic values of the electron’s wavenumber and mass are all represented in this equation by the variables $k$ and $m$ [8].

The experimental findings made by Loudon [4] and the queries we raised before served as the main inspirations for our decision to use NiO for our computational work. Due to the presence of heavier elements (La and As) in this
compound, we chose the second antiferromagnetic material, LaMnAsO, for our calculations. As a result, we anticipate stronger TDS, which may make it more difficult to detect a weak Bragg spot. By doing so, we are able to qualitatively compare whether it is possible to find the magnetic Bragg spot in studies using a variety of different material compositions and experimental setups. We present the multislice findings diffractional patterns for static model computation exhibiting antiferromagnetic Bragg spots in both compounds in Fig. 1.

To conclude, our calculations reveal that it is certainly conceivable to see the $\frac{1}{2}(111)$ antiferromagnetic Bragg spot in NiO at room temperature, as Loudon demonstrated experimentally. We have established that the thermal diffuse scattering intensity is much lower than the antiferromagnetic Bragg spot intensity. Additionally, there is good agreement between our estimates and the measured thickness dependence of the magnetic Bragg spot intensity. Our calculations for LaMnAsO, which contains heavier elements than NiO, showed that it can be difficult to find the magnetic Bragg spots in systems with substantial thermal diffuse scattering. It might be necessary for such systems to operate at lower temperatures and/or to carry out data collecting for a prolonged period of time with a sufficiently strong beam current.

The findings also point to the necessity of using the right acceleration voltage, which for the materials under study here is determined to be 300 kV. It will be possible to employ TEM for high-resolution detection of complicated magnetic ordering by using the simulation techniques that have been given. An extended presentation of the results is available in Ref. [9].

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