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Element-specific magnetic states in Fe_4N studied by XMCD

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The 3*d* transition-metal-based nitride alloy films, such as Fe_4N and Mn_4N, exhibit unique and exceptional properties in spintronics applications, including the anomalous Hall and Nernst effects, anisotropic magneto-resistance, and spin current generation [1]. They form an anti-perovskite-type cubic crystal structure as a unit cell, where nitrogen atom stably occupies the body-centered site of the host face-centered cubic structure formed by Fe and Mn atoms. The hybridization between Fe 3*d* and N 2*p* states, which enhances the chemical covalent bonds compared to oxide cases, leads to unique band structures near the Fermi level that contribute to the negative spin polarization of −0.6 in Fe_4N, resulting in the inverse tunneling magnetoresistance effect [2].

One of the recent significant issues in nitride spintronics focuses on the detailed growth conditions for Fe_4N, including substrate choice, growth temperature, and various annealing temperatures after deposition. To date, several substrates such as MgO (001), $SrTiO_3$, $LaAlO_3$, and Cu have been utilized due to their lattice matching with Fe_4N. Among these, the MgO substrate yields the highest spin polarization. Therefore, in this study, we aim to investigate the dependence of annealing temperature on Fe_4N thin films using x-ray magnetic circular dichroism (XMCD) to understand the relationship between magnetic properties and magneto-transport characteristics.

30-nmn-thick Fe_4N films were grown by magnetron sputtering with nitrogen reactive sputtering at substrate temperatures (T_{sub}) of 450, 550, and 650 °C on MgO (001) substrates. The films were capped with a 1-nm-thick Al layer to prevent surface oxidation. XAS and XMCD spectroscopies were conducted at BL-7A in the Photon Factory at the High-Energy Accelerator Research Organization (KEK-PF). For the measurements, the photon helicity was fixed, and a magnetic field of ±1.2 T was applied parallel to the incident polarized soft x-ray beam to obtain signals defined as μ+ and μ− XAS spectra. XMCD is defined as the difference between μ+ and μ− XAS spectra. The total electron yield mode was adopted, and all measurements were performed at room temperature. The XAS and XMCD measurement geometries were set to oblique incidence, ensuring that both the photon helicity axis and the magnetic field were parallel, which enabled the measurement of absorption processes involving the in-plane components of the spin and orbital magnetic moments.

Figure 1 presents the Fe *L*-edge XAS and XMCD of Fe_4N films measured at room temperature for T_{sub} = 450 and 650 °C. In both instances, distinct metallic line shapes were observed. In the highly ordered case, the XAS line shapes display shoulder structures, whereas the 450 °C case reveals a broader structure. This suggests that the two types of Fe sites with Fe-Fe and Fe-N bonds can be clearly differentiated. The line shapes align with previous reports of XMCD in Fe_4N [3]. The local charge transferred in Fe-N bonds indicates a chemical shift towards the high-energy side. Although the degree of order varies between the two cases, the XMCD intensities remain nearly identical, implying that the total magnetic moments are relatively insensitive to atomic ordering.

To investigate the induced magnetic moments at the nitrogen sites, XAS and XMCD at the N *K*-edge were conducted. XAS reveals structures from the nitrogen associated with certain chemical bonds. Minor differences of 3% between μ+ and μ- normalized XAS can be identified as XMCD signals. We emphasize that a systematic investigation of ordering dependence facilitates an understanding of the behavior of nitrogen atoms in Fe_4N. These results indicate that either the Fe_4N phase or another phase of Fe-N compounds may coexist in the film at 450 °C.

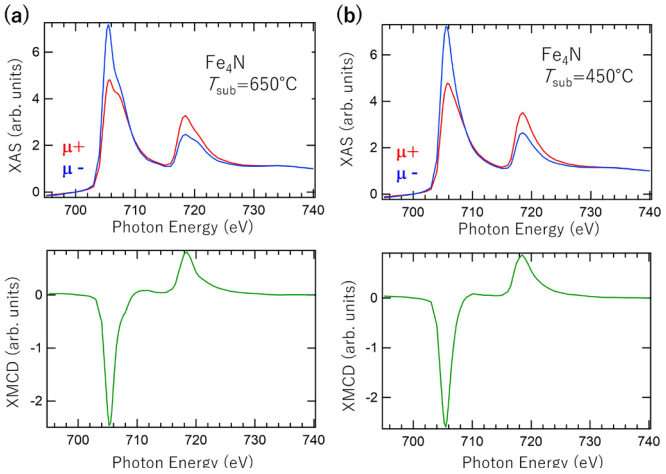

Fig. 1, Fe *L*-edge XAS and XMCD for Fe_4N films grown at (a) 650°C and (b) 450°C.

References

[1] S. Isogami, and Y. K. Takahashi, Adv. Electron. Mater. **9**, 2200515 (2023).
[2] A. Sakuma, J. Phys. Soc. Jpn. **60**, 2007 (1991).
[3] K. Ito et al., J. Appl. Phys. **117**, 193906 (2015).

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