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Element-Specific Electronic Structure and Magnetic Coupling in $NiFe_2O_4/BaTiO_3$ Composites Probed by XAS and XMCD

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1 Introduction

Multiferroic composites, combining ferroelectric and magnetic phases, have significant interest for magnetoelectric (ME) coupling through strain-mediated interactions. These materials are very useful for advanced applications in spintronics, magnetic sensors, energy harvesters, microwave devices, and non-volatile memory. Nickel ferrite ($NiFe_2O_4$)/Barium titanate ($BaTiO_3$) composite is one of the most thoroughly studied multiferroic systems due to its strong structural compatibility, chemical stability, and synergistic ferroic properties [1].

$NiFe_2O_4$ is an inverse spinel ferrite characterized by ferrimagnetic ordering, high electrical resistivity, moderate saturation magnetization, low eddy current loss, and strong chemical and thermal durability. These make it a compelling magnetic phase for multifunctional composites [1-7]. $BaTiO_3$ is a lead-free ferroelectric perovskite characterized by strong dielectric permittivity, spontaneous polarization, and superior piezoelectric characteristics. Combining these two phases allows for the mechanical transmission of the piezoelectric strain produced in $BaTiO_3$ to $NiFe_2O_4$ across the contact, producing magnetoelectric coupling [1]. The efficacy of this coupling is heavily influenced by parameters including phase composition, particle dispersion, interfacial bonding, and microstructure. Consequently, adjusting the $BaTiO_3$ content in $NiFe_2O_4/BaTiO_3$ composites is important for customizing their magnetic and magnetoelectric properties. This study produced $NiFe_2O_4/BaTiO_3$ composites by a solution dispersion method and thoroughly studied their electronic and magnetic properties to elucidate the impact of $BaTiO_3$ incorporation on the multifunctional performance of the composites.

2 Experiment

$NiFe_2O_4/BaTiO_3$ composites were prepared by a solution dispersion method. The requisite quantity of $BaTiO_3$ was ultrasonically dispersed in ethanol for 60 minutes, then $NiFe_2O_4$ was independently dispersed in ethanol using magnetic stirring for 1 hour. The $BaTiO_3$ suspension was thereafter introduced systematically to the $NiFe_2O_4$ dispersion while maintaining continuous agitation to achieve uniform mixing. The heterogeneous solution was subjected to heating at 100–110 °C with continuous agitation until the solvent was completely evaporated, resulting in a homogeneously blended composite powder. The resultant powder was finely milled to produce $NiFe_2O_4/BaTiO_3$ composite samples for further characterizations.

X-ray Absorption Spectroscopy (XAS) and X-ray Magnetic Circular Dichroism (XMCD) were used to study the electronic and magnetic properties of elements at the Ni and Fe $L_{2,3}$ edges on the undulator beamline BL-16A at the Photon Factory (KEK), Japan. The XMCD spectra were obtained by measuring the difference between the absorption coefficients for photons with two different circularly polarized helicities, denoted by μ^+ and μ^-. Measurements were performed in total electron yield (TEY) mode under ultra-high vacuum (~10^{-9} Torr) at room temperature (300 K). Circularly polarized X-rays with a polarization degree of about 87 ± 4% and an energy resolution of E/ΔE > 10,000 were applied. The XAS and XMCD spectra were obtained at a magnetic field of 1 T.

3 Results and Discussion

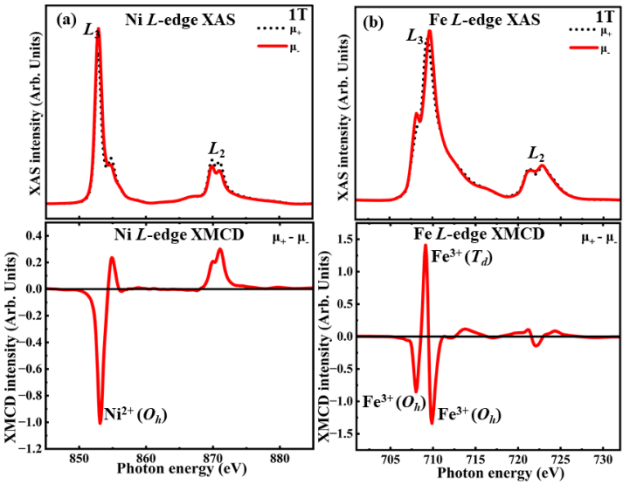

Figure 1: Ni and Fe $L_{2,3}$-edge XAS and XMCD of $NiFe_2O_4/BaTiO_3$ composites.

Figure 1(a) shows the Ni *L*-edge XAS and XMCD spectra of $NiFe_2O_4/BaTiO_3$ composites, providing element-specific electronic and magnetic information. XAS spectra exhibited distinct L_3 and L_2 absorption edges, which correspond to the $2p_{3/2} \rightarrow 3d$ and $2p_{1/2} \rightarrow 3d$ transitions, confirming the existence of Ni^{2+} ions. XMCD spectrum exhibited a strong dichroic signal, indicating the magnetic influence of octahedrally coordinated Ni^{2+} (O_h) ions on the ferrimagnetic ordering. **Figure 1(b**) displays the Fe *L*-edge XAS and XMCD spectra. XAS spectra exhibited distinct L_3 and L_2 characteristics of Fe^{3+} ions. XMCD spectrum revealed distinct positive and n
egative peaks, indicative of Fe^{3+} ions occupying T_d as well as O_h sites, thereby confirming the antiparallel spin alignment, which is the basic feature of the inverse spinel $NiFe_2O_4$ structure and its ferrimagnetic properties [1,3,7]. When $BaTiO_3$ is added to $NiFe_2O_4$ near the overlap of the two helicities signals indicates a stable electronic structure with slight valence change. The spectra demonstrated pronounced element-specific magnetic ordering in $NiFe_2O_4/BaTiO_3$ composites, with $BaTiO_3$ not disrupting the ferrimagnetic structure.

References

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