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Polarons induced electronic transport, dielectric relaxation and magnetodielectric coupling in spin frustrated Ba_2FeWO_6

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ABSTRACT

Mixed valent double perovskite Ba_2FeWO_6, with tetragonal crystal structure, synthesized in a highly controlled reducing atmosphere, shows antiferromagnetic transition at $T_N = 19$ K. A cluster glass-like transition is observed around 30 K arising from the competing interactions between inhomogeneous magnetic states. The structural distortion leads to the formation of polarons that are not contributing to DC conduction below charge ordering temperature, $T_{CO} = 279$ K. Above T_{CO}, small polarons will start to hop by exploiting thermal energy and participate in the conduction mechanism. The polarons are also responsible for the dielectric relaxor behavior, in which the dielectric relaxation time follows non-linearity in temperature as proposed by Fulcher. The material also exhibits a small room temperature magnetoresistance of 1.7% at 90 kOe. An intrinsic magnetodielectric coupling of ~4% near room temperature and at lower temperatures, as well as an extrinsic magnetodielectric coupling change from +4% to −6% at around 210 K are reported.

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1. Introduction

Perovskite materials are oxides of transition metals (general formula ABO_3) with interesting physical properties. The double perovskites (DPs), with the general formula $A_2BB'O_6$, (A being alkaline earth metals or rare earth metals and B, B′ being transition metals), exhibit diverse properties such as insulating/dielectric, metallic/half metallic, antiferromagnetic (AFM)/ferromagnetic (FM)/ferrimagnetic (FiM), intergrain tunneling type magnetoresistance, magnetodielectric, and multiferroics [1–3]. Different electronic and magnetic properties of DPs are due to the imperfections associated with B/B′ ordering, oxidation states of B and B′ cations, anti-phase boundaries, B–O–B′ exchange interaction, and lattice distortion incorporated with DP structure [4,5]. In the ordered DP, BO_6 and $B'O_6$ octahedra are alternatively arranged in two interleaving FCC sublattices. Due to the mismatch in size between A and B/B′ cations and the mismatch in charge, size and electronic configuration of the B/B′ cations, the octahedra undergo tilting and distortion to achieve the most energetically favorable structure that may cause to produce polarons in the lattice [4].

The DP material Ba_2FeWO_6 (BFWO) was first reported with cubic Fm3 m crystal structure [6,7]. Later Azad et al. studied the material and observed a tetragonal I4/m crystal structure. They observed AFM structure with Néel temperature, T_N~20 K based on neutron powder diffraction studies and a Reverse Monte Carlo modeling [8,9]. Rammeh et al. reported BFWO as cubic Fm3 m structure with a spontaneous magnetization of 0.152 μ_B/Fe at 5 K which is lower than theoretically expected saturation magnetization due to the inhomogeneities associated with the Fe/W site disorder [10]. The charge distribution in B site cations was Fe^{2+} and W^{6+} when prepared in nitrogen atmosphere [9]. If prepared in partially reducing atmosphere, we may be able to obtain mixed oxidation states of B/B′ cations and thereby alter the magnetic and transport properties by introducing inhomogeneities and distortion. Magnetic properties in DP was reported to be arising from various interactions like hybridization driven AFM/FiM, super exchange driven microscopically originated FM/FiM, intra-atomic

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exchange interaction, and hybridization driven negative spin splitting [11–13].

In the present investigation, a controlled reducing atmosphere was used for the synthesis of BFWO, which helped the material to achieve mixed oxidation states and the allied imperfections. The superexchange interaction between Fe^{2+} (t_{2g}^4, e_g^2)–W^{6+} (t_{2g}^0, e_g^0) and Fe^{3+} (t_{2g}^3, e_g^2)–W^{5+} (t_{2g}^1, e_g^0) will be FM. Also, there will be interplanar AFM coupling of the FM planes. But the mis-site effect contributes to the incompatible exchange interactions viz. Fe^{2+}–O–Fe^{2+}, Fe^{2+}–O–Fe^{3+}, Fe^{2+}–O–W^{5+}, Fe^{3+}–O–Fe^{3+}, Fe^{3+}–O–W^{6+}, W^{5+}–O–W^{5+}, W^{6+}–O–W^{6+} and W^{5+}–O–W^{6+} and their complicated couplings which may give rise to complicated magnetism and magnetotransport properties in the material [14]. The present work reports a detailed experimental study about the presence of spin frustration, along with electronic transport properties, magnetoresistance, dielectric relaxation and magnetodielectric coupling behavior in BFWO.

2. Synthesis and characterization

Polycrystalline BFWO was prepared by the standard solid-state ceramic route with thermal treatment in controlled reducing atmosphere. Stoichiometric ratio of precursor mixture of high purity carbonates and oxides ($2\ BaCO_3 + 1/2\ Fe_2O_3 + WO_3$) were mixed for two hours uniformly in a medium of de-ionized water and kept inside a hot air oven at 80 °C for one day. The dried mixture was ground well and heated to 800 °C for 12 h in air. The pelletized material was kept in an evacuated tube furnace and blowed with high purity H_2:Ar mixture gas mixed in 1:19 molar ratio. The material is then sintered at 1200 °C for 6 h, and cooled to 200 °C under the controlled reducing mixture gas atmosphere, and then air cooled to room temperature. The precursor cations Fe^{3+} and W^{6+} in the reducing thermal atmosphere self-tune the oxidation state between Fe^{2+}–Fe^3 and W^{5+}– W^{6+} to form the preferred ordered DP structure of BFWO. Powder X-ray Diffraction (XRD) pattern of the ground pellet at room temperature was taken by using PANalytical X'Pert Pro Powder X-ray diffractometer with a Ni filtered Cu Kα radiation (λ = 1.5405 Å). The structural refinement was done by GSAS using EXPGUI platform [15]. The crystallographic structure is framed using CrystalMaker® [16]. X-ray photoelectron spectroscopy (XPS) was done by using Thermo Scientific X-ray photoelectron spectrometer, MULTILAB 2000 Base system with X-Ray, Auger and ISS attachments. The Fe and W spectra obtained are fitted using the XPS peak Fit software. DC magnetization studies were done by using a Vibrating Sample Magnetometer (VSM) option attached to a Physical Property Measurement System (PPMS) (The Quantum Design, Dynacool). The AC susceptibility studies were carried out using a Superconducting Quantum Interference Device (SQUID-Quantum Design) system. DC resistivity was obtained by the two-probe method using the Electrical Transport Option (ETO) attached to the PPMS. The dielectric measurements have been performed with an LCR meter (Agilent E4980A) which has been compensated for the coaxial cable length and calibrated for open and short circuit. Pure silver-plated pellet of thickness 1.61 mm and diameter 10.28 mm, connected as a parallel plate capacitor, is inserted into the cold head of the closed cycle Cryocooler (ARS-2HV). The magnetic field was applied to the capacitor connection by inserting them into the middle of a hollow cylindrical Neo magnet.

3. Results and discussions

3.1. Structural analysis (X-ray diffraction and X-ray photoelectron spectroscopy)

The theoretical tolerance factor calculated using Shannon radii [17] is 1.02 for BFWO and that observed $f_{obs} = d_{A-O}/\sqrt{2}\left(\left\langle d_{B-O}\right\rangle\right)$ from the refined results is 0.999. For the DP family, $f < 0.97$ suggests either a monoclinic or orthorhombic structure, $0.97 < f < 1$ suggests I 4/m tetragonal structure, $1 < f < 1.05$ suggests Fm 3 m cubic structure and $f > 1.05$ suggests hexagonal structure [18]. Rietveld refinements of XRD pattern is

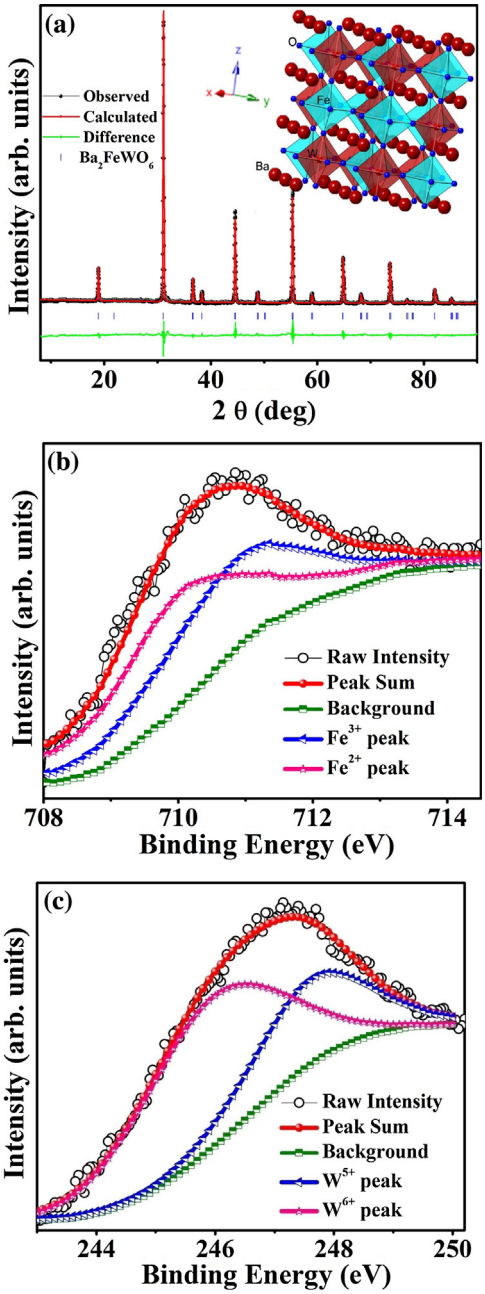

Fig. 1. (a) Observed, calculated and the difference XRD pattern of Ba_2FeWO_6 obtained from Rietveld refinement. The bar marks represent the possible Bragg reflections. Inset: the crystallographic structure framed using CrystalMaker®. (b) XPS spectra of Fe $2p_{3/2}$ in Ba_2FeWO_6 along with the fitted curves. (c) XPS spectra of Fe $W4d_{5/2}$ in Ba_2FeWO_6 along with the fitted curves.

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