Journal of Magnetism and Magnetic Materials 320 (2008) 2591–2595

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Magnetic properties, electronic structure, and optical properties of the filled skutterudite $BaFe_4Sb_{12}$

Zheng-ze Cheng [a,b,*], Ze Cheng [a], Ming-hu Wu [c]

[a] Department of Physics, Huazhong University of Science and Technology, Wuhan 430074, China
[b] Department of Physics, Xian Ning College, Xian Ning, 437005, China
[c] Department of Chemistry, Xian Ning College, Xian Ning, 437005, China

ARTICLE INFO

Article history:
Received 19 March 2008
Received in revised form
23 April 2008
Available online 18 May 2008

PACS:
71.20.−b
78.20.−e
71.15.Ap

Keywords:
First-principle
Filled skutterudite
Optical property

ABSTRACT

The magnetic properties, electronic structure, and optical properties of the filled skutterudite $BaFe_4Sb_{12}$ are calculated by the first-principles full-potential linearized augmented plane wave (FPLAPW) plus local orbital method. It is found that the local spin density approximation (LSDA) method appears more accurate than the generalized gradient approximation (GGA) method in calculating the electronic structures and optical properties of this compound. Furthermore, our calculated lattice constant and spin magnetic moments with the LSDA method are in overall better agreement with experiment. In contrast with recent experiment, our calculations are in good agreement with experimental reflectivity spectra and optical conductivity spectrum.

1. Introduction

Ternary compounds RM_4X_{12} (R = rare earth elements; M = Fe,Ru,Os; X = P,As,Sb) with the filled skutterudite structure (space group Im3) exhibit a wide variety of physical properties. Previous studies have shown that their physical properties are in large part determined by the M atoms. A variety of interesting physical phenomena, including superconductivity, magnetic order, small hybridization gap semiconductivity (also known as "Kondo insulator" behavior), valence fluctuation and heavy fermion behavior, non-Fermi liquid behavior, and metal–insulator transition have been observed among the various members of the filled skutterudites [1–11]. On the other hand, these compounds attracted renewed attention due to their potential application as thermoelectric materials [12–14].

Recently, the optical properties of filled skutterudite compounds have been of interest to experimentalists and theorists alike. Danebrock et al. have studied the magnetic properties of the nine title compounds by magnetic susceptibility measurements with a SQUID magnetometer between 2 and 300 K [4]. Leithe-Jasper et al. have investigated the synthesis, chemical, structural, and magnetic properties of alkali-metal compounds with filled skutterudite structure, $NaFe_4Sb_{12}$ and KFe_4Sb_{12} [15]. Magnetic and thermoelectric properties of the skutterudites AT_4Sb_{12} filled with non-magnetic atoms, A = Ca, Sr, Ba, and La, are found to depend strongly on the transition-metal elements, T = Fe, Ru, and Os by Takabatake et al. [16] Magnetic properties of the alkaline earth-filled skutterudite AFe_4Sb_{12} (A = Ca, Sr, Ba), which are located in a proximity to a ferromagnetic instability, have been investigated by the high-field magnetization measurements in a pulsed magnetic field by Yoshii et al. [17] Sichelschmidt et al. have investigated optical properties of the filled skutterudites AFe4Sb12 with divalent cations A = Yb, Ca, Ba [18]. Krishnamurthy et al. have combined X-ray magnetic circular dichroism spectroscopy at Fe $L_{2,3}$ edges, at Eu $M_{4,5}$ edges, X-ray absorption spectroscopy (XAS) investigation of Eu valence, and local spin density calculations [19].

The aim of this paper is to present the results of a theoretical investigation of magnetic properties, electronic structure, and optical properties for filled skutterudite compounds $BaFe_4Sb_{12}$, based on the full-potential linearized augmented plane wave (FPLAPW) method.

2. Computational details

The presented calculations were performed within the generalized gradient approximation (GGA) to the density functional theory, using the FLAPW method and its extended version Wien2k [20,21]. In this method no shape approximation on either

* Tel.: +86 13451080298.
E-mail address: czz8986@126.com (Z.-z. Cheng).

0304-8853/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2008.05.017

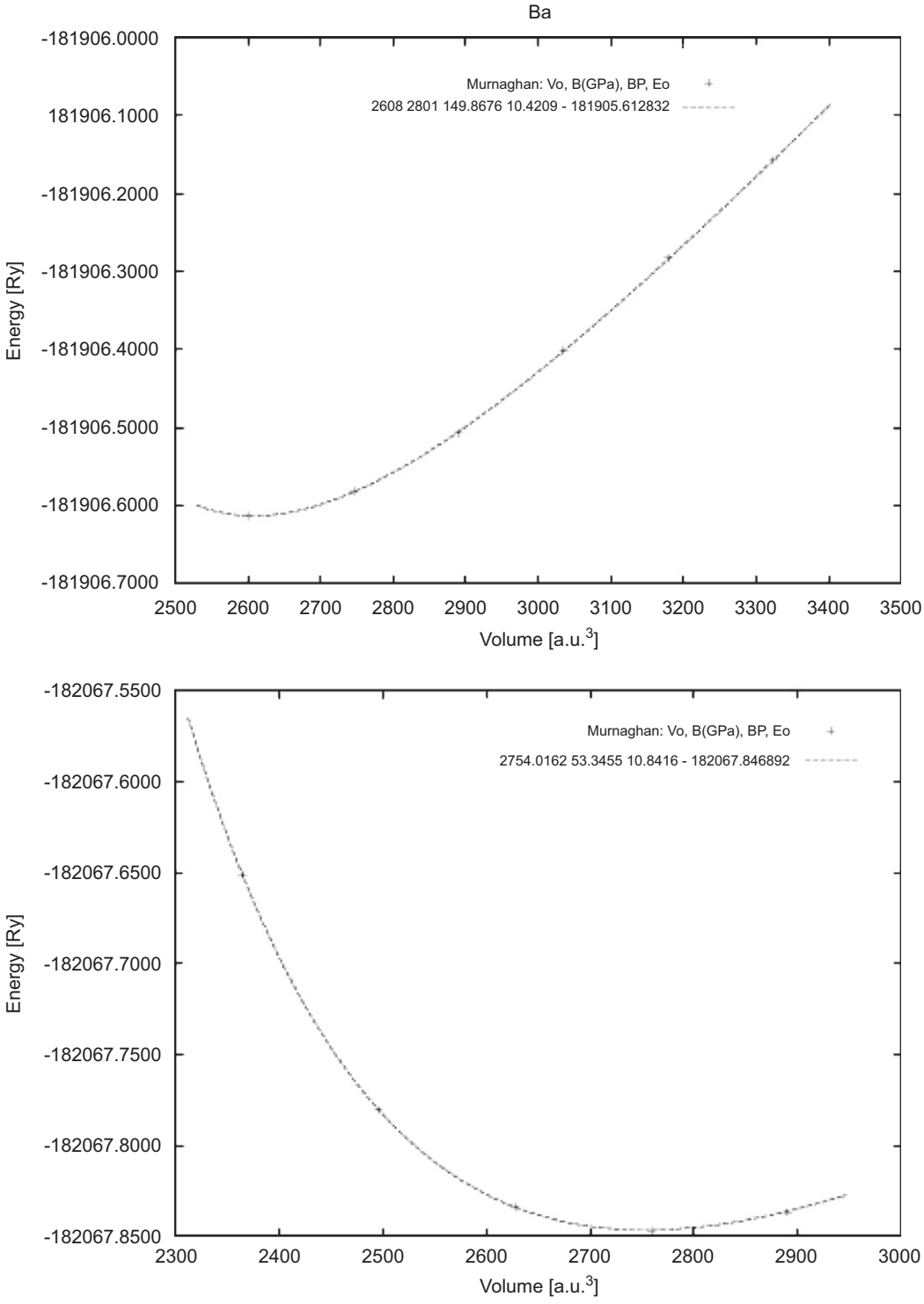

Fig. 1. Total energy versus volume curves for $BaFe_4Sb_{12}$ within LSDA (left panel) and GGA (right panel).

potential or the electronic charge density is made. We use Wien2k implementation of the method, which allows the inclusion of local orbits in the basis, improving upon linearization and making possible a consistent treatment of semicore and valence states in one energy window, hence ensuring proper orthogonality. In the FLAPW method, the unit cell is divided into two parts: (1) non-overlapping atomic spheres (centered at the atomic sites) and (2) an interstitial region. The lattice parameters we used are 9.1865 Å (experimental results 9.2000 Å) [22]. The atomic sphere radii 2.5, 2.5, and 2.26 a.u. are used for Ba, Fe, Sb, respectively, in the calculations. The convergence parameter RK_{max} ($R_{mt}{}^{*}K_{max}$, where K_{max} is the plane-wave cut-off and R_{mt} is the smallest of all atomic sphere radii), which controls the size of the basis sets in these calculations, is set to be 8.0. We have used $10\times10\times10$ meshes, which represent 1000 k-points in the first Brillouin zone.

The dielectric tensor has been calculated from Wien2k code. Using the $\varepsilon(\omega)$ (where $\varepsilon(\omega)=\varepsilon_1(\omega)+i\varepsilon_2(\omega)$), as output, we calculated various optical constants [23] such as the spectral reflectivity $R(\omega)$ with a simple independent program.