April 1994

Electronic structure and magneto-optical properties of MnBi and MnBiAl

Sitaram Jaswal
University of Nebraska, sjaswal1@unl.edu

J. X. Shen
University of Nebraska - Lincoln

Roger D. Kirby
University of Nebraska-Lincoln, rkirby1@unl.edu

David J. Sellmyer
University of Nebraska-Lincoln, dsellmyer@unl.edu

Electronic structure and magneto-optical properties of MnBi and MnBiAl

S. S. Jaswal, J. X. Shen, R. D. Kirby, and D. J. Sellmyer
Behlen Laboratory of Physics and Center for Materials Research and Analysis, University of Nebraska, Lincoln, Nebraska 68588-0111

MnBiAl films are of considerable current interest for possible magneto-optical applications because of their perpendicular anisotropy, large polar Kerr rotation, reduced grain size, and structural stability. We report here experimental and theoretical studies of the effect of Al alloying on the electronic structure and magneto-optical properties of MnBi. Our measured spectral dependencies of the polar Kerr rotation in the two systems are similar. We carried out relativistic self-consistent spin-polarized electronic structure calculations on MnBi and MnBiAl, and the calculated densities of states are in good agreement with available x-ray photoemission spectroscopy data for both materials. The spin magnetic moment of Mn increases and the overall orbital moment decreases with the addition of Al to MnBi. The latter effect implies the lowering of spin-orbit interactions upon alloying MnBi with Al, and the two effects together suggest that the spectral dependencies of the Kerr rotation should be similar, as is observed experimentally.

MnBi has been known for several decades as a material with a large Kerr rotation. Recently, efforts have been made to improve its magneto-optical properties by alloying with other metals such as Al.[1–4] The electronic structure of MnBi has been calculated by Coehoorn and deGroot,[5] and a cluster study on the electronic and magnetic properties of MnBi and MnBiAl has been reported.[6] We report here theoretical and experimental studies of the electronic structure and magneto-optical properties of MnBi and MnBiAl.

MnBi crystal is hexagonal with four atoms per unit cell and the space group symmetry is $P6_3/mmc$ with sites a and c occupied. MnBiAl also has the same structure, and the distance between the hexagonal planes remains essentially unchanged upon alloying MnBi with Al. Unfortunately, little information is available on the change in the a lattice constant with the addition of Al. Also, the sites occupied by the Al atoms are unknown, but the interstitial sites in the hexagonal NiAs structure are thought to be the most likely candidates. In the present calculations we assume MnBiAl to be an ordered structure with all the interstitial (d) sites occupied by Al atoms. We had to make the a lattice constant of MnBiAl 15% larger than that of MnBi to stabilize the electronic structure calculations.

The relativistic self-consistent spin-polarized electronic structure calculations are based on the linear muffin-tin orbitals method (LMTO) in the local density approximation.[7] MnBi and MnBiAl have four and six atoms per unit cell, respectively. With s, p, and d as basis functions, the relativistic Hamiltonian gives 72×72 and 108×108 matrices for MnBi and MnBiAl, respectively.

The results for total and partial spin-polarized densities of states (DOS) for MnBi are shown in Fig. 1. The Mn $3d$ states dominate the spectrum with a peak around 3 eV below the Fermi level in the occupied states. The magneto-optical properties of MnBi are primarily determined by transitions between Mn d and Bi p states. The present results are similar to those of Coehoorn and deGroot[5] based on the augmented spherical wave method.

The MnBiAl DOS, plotted in Fig. 2, are qualitatively similar to those of MnBi. The differences in the two spectra arise because of the additional hybridization mainly between the s-p states of Bi and Al in MnBiAl. The calculated DOS for the two systems are in good agreement with the x-ray photoemission spectroscopy data of Wang *et al.*[2]

The calculated magnetic moments in MnBi and MnBiAl are listed in Table I. The Mn moment in MnBiAl is about 5% larger than that in MnBi, whereas the induced moment on Bi goes down. This is due to the reduced hybridization between Mn and Bi states, which results from the hybridization of the Bi and Al states mentioned in the preceding paragraph.

The Kerr rotation in the two systems is primarily due to the magnetism of Mn and the spin-orbit interactions of Bi.[8]

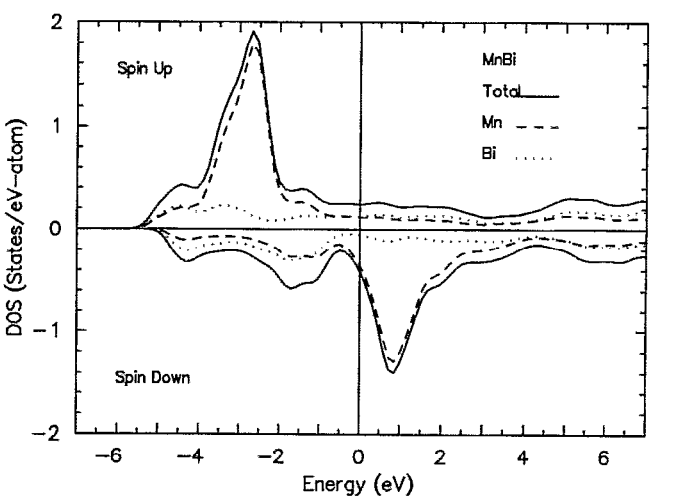

FIG. 1. Total and partial spin-polarized densities of states of MnBi.

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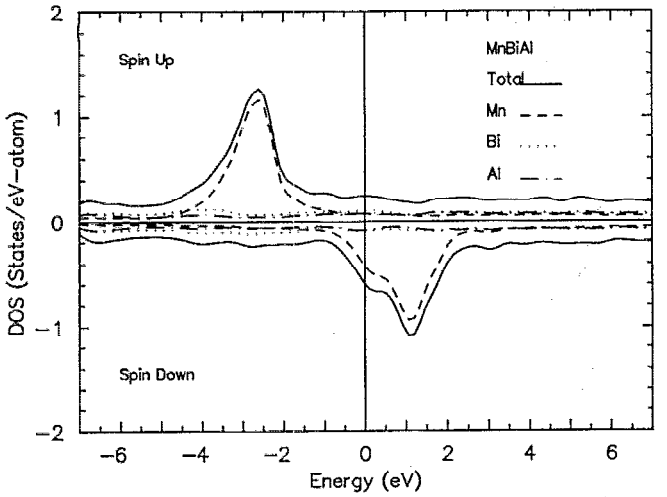

FIG. 2. Total and partial spin-polarized densities of states of MnBiAl.

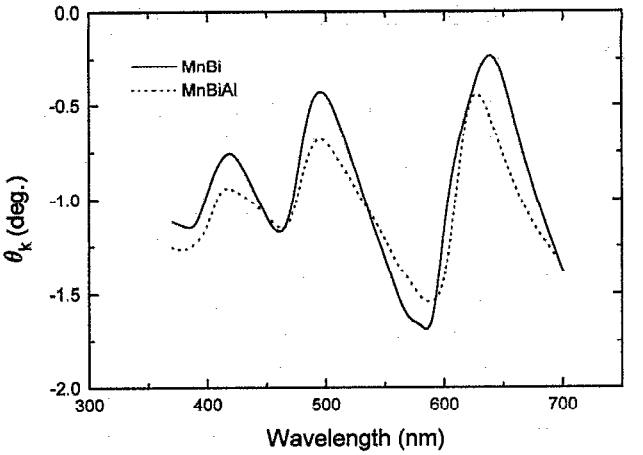

FIG. 3. Measured polar Kerr rotation as a function of the wavelength in MnBi and MnBiAl. The oscillations in the rotation are due to optical interference effects in the SiO_x overcoat. The nominal compositions were $Mn_1Bi_{0.8}$ and $Mn_1Bi_{0.8}Al_{0.4}$.

We have seen above that the addition of Al to MnBi enhances the Mn magnetic moment. To study the effect of Al on spin-orbit interactions, we have calculated the orbital moments in the two systems, which are given by

$$l_z = \sum_{\sigma l m_l} n^{\sigma}_{m_l} m_l ,$$

where m_l is the magnetic quantum number corresponding to the orbital quantum number l and $n^{\sigma}_{m_l}$ is the occupied number density for spin σ.[9] The calculated values of the orbital moment (in μ_B) for MnBi and MnBiAl are 0.129 and 0.057 per unit cell, respectively. Thus the addition of Al to MnBi lowers the spin-orbit interactions due to the hybridization of Al and Bi states.

The present calculations are based on 33.3 at. % Al in MnBiAl, while the experimental results are for Al concentrations of typically 15 at. %. Thus the increase in the magnetic moment of Mn and the decrease in spin-orbit interactions for the experimental concentrations of Al in MnBi are expected to be small. Since the changes in the magnetic moments and spin-orbit interactions due to aluminization of MnBi have opposite effects on the Kerr rotation, we expect Kerr spectra of MnBi and MnBiAl to be quantitatively similar.

TABLE I. Magnetic moments (in μ_B) for different sites in MnBi and MnBiAl.

	MnBi		MnBiAl
	Present calculations	Ref. 5	Present calculations
Mn	3.78	3.61	4.08
Bi	−0.20	−0.09	−0.01
Al			−0.02

Our experimental results for the Kerr rotation as a function of wavelength are plotted in Fig. 3. The oscillations in the data are due to multiple reflections from the ~150 nm thick SiO_x overcoat. Taking the average values of these oscillations, we note that the Kerr rotations for MnBi and MnBiAl are similar, in agreement with our theoretical predictions. Our experimental results are also in agreement with those of Di *et al.*[4] The experimental results of Wang *et al.*[1,2] are somewhat similar to the above data except for a low value of 0.7° for MnBi at 633 nm.

In conclusion, the electronic structure calculations predict that the Kerr rotations in MnBi and MnBiAl should be similar, which is in agreement with the experimental data.

This work is supported by the National Science Foundation under Grant No. DMR-9222976, ARPA Grant No. MDA 972-93-1-0009 to the National Storage Industry Consortium, the NSF Experimental Program to Stimulate Competitive Research, and the National Supercomputing Facility at Cornell University. We are grateful to Hans Skriver for the copies of relativistic LMTO programs.

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