

Examination of the Mn Local Structure of $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ and $\text{Ga}_{1-x-y}\text{Mn}_x\text{Be}_y\text{As}$ Using EXAFS

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Introduction

One of the recent topics of interest in materials science/condensed matter physics has been a group of systems known as the “spintronic” materials. These materials are semiconductors with magnetic moments, thus coupling the electrical properties with magnetic moments. The materials of this investigation, $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ and $\text{Ga}_{1-x-y}\text{Mn}_x\text{Be}_y\text{As}$, are both from this group. The Mn added to the GaAs provides magnetic moments and holes, while the Be in the second group of materials adds only holes. The interest in these materials increased with the announcement of Curie temperature as high as 110 K [1,2]. The ferromagnetic nature of these materials is believed to occur due to an RKKY-like interaction between Mn ions, which is moderated by the holes in the materials. As such, two directions to improve the T_c are to increase the amount of Mn participation the ferromagnetic interaction, (either by increasing the concentration of Mn or by increasing the likelihood of contribution to the interaction by a given Mn) or to increase the number of holes which moderate the interaction. Annealing the $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ and adding Be to it have been examined as methods to improve one of these possibilities, respectively.

Methods and Materials

X-ray absorption fine-structure spectroscopy (XAFS) is a technique which provides local structural information in the area around atoms of a specific element. This technique does not differentiate between atoms of the same element in different structural environments, so if the atoms of the element being studied are in more than one structural environment, the signals from each environment are combined. This method is well suited for examining these types of materials as the Mn edge energy is well separated in energy from the edges of the other elements in the samples; Ga, As, and Be.

The samples were grown at Notre Dame in a Riber 32 R&D MBE system. The details of this growth can be found elsewhere [3,4]. The $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ samples were grown with $x = 0.01, 0.02, 0.05,$ and 0.08 . The $\text{Ga}_{1-x}\text{Mn}_x\text{Be}_y\text{As}$ samples were grown with $x = 0.05$ and a range of values for y from 0.0 to 0.11 . The Be codoped samples will hereto be referred to by the Be cell temperature during grown rather than the percentage of Be grown. Table 1 shows the correlation between cell temperature and Be concentration.

The experiments were conducted at MRCAT, sector 10-ID. The first harmonic of the undulator was used with a 0.1 KeV taper and a Si 111 monochromator to provide a beam at the energy of the Mn edge, 6.539 KeV. A glass

Be Cell Temperature	Be Content, y
0	0
960	0.01
1038	0.03
1050	0.048
1060	0.08
1070	0.11

Table 1

harmonic rejection mirror was used to remove higher order harmonics from the beam, so that they would not contaminate the data. Due to the highly crystalline nature of the MBE-grown samples, Bragg reflection posed a threat to data collection. In order to remove this threat, the x-ray fluorescence was measured in grazing incidence geometry using a 13-element solid state detector. Following data reduction using standard methods, the data was fitting using the IFEFFIT program [5]. Several models were used to simulate the structures, namely Mn_{Ga} (a substitutional model), $\text{Mn}_{\text{I-As}}$ (an interstitial Mn coordinated by 4 As), $\text{Mn}_{\text{I-Ga}}$ (an interstitial Mn coordinated by 4 Ga), Mn_{Hex} (an interstitial Mn coordinated by 3 As and 3 Ga), MnAs (a possible precipitate in the materials), and Mn metal (a possible precipitate. IFEFFIT was then used to fit the theoretical scattering paths from the models (generated using FEFF 8.2) to the data [6, 7].

Results

The χ data from the $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ was Fourier transformed from 2.4 to 12.5 and the magnitude of this transform is shown in Fig. 1. This data shows the similarities in the nearest neighbor local structure around the Mn as a function of Mn concentration. The $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ materials were simulated using the models listed earlier. All four concentrations of Mn were found to be well simulated using the first 3 non-multiple scattering paths of the Mn_{Ga} model.

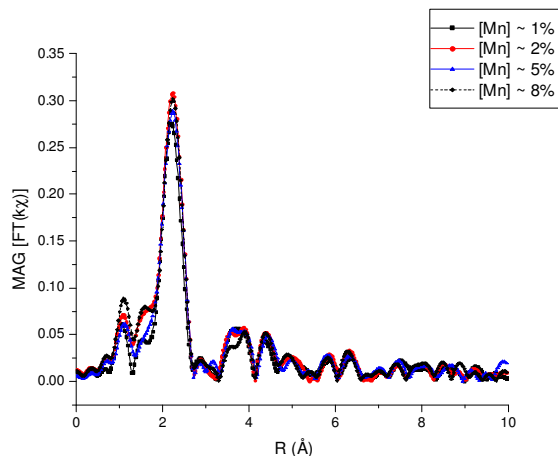


Fig. 1

The $\text{Ga}_{1-x-y}\text{Mn}_x\text{Be}_y\text{As}$ materials were Fourier transformed from 2.4 to 12.5, and the magnitude of these Fourier transforms are shown in Fig. 2. This figure shows that the magnitude of the first shell scattering decreases as the concentration of Be increases. This decrease could be caused by several things. The first possibility is that the coordination number of the Mn has changed. The second possibility is that the decrease in the first shell scattering amplitude is caused by an increase in the Debye-

Waller factor. The third possibility is that the Mn is no longer in a single local environment and the signal from the various local environments interferes destructively.

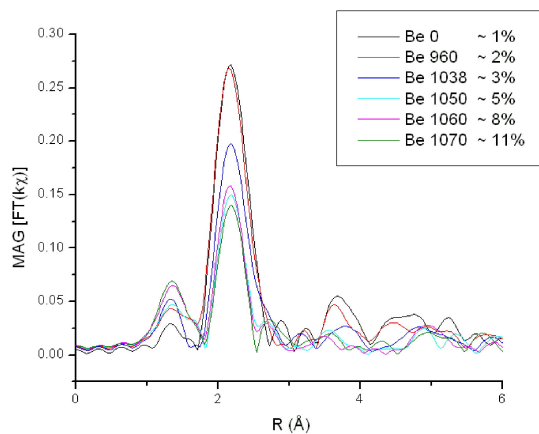


Fig. 2

The first possibility, a change in the coordination number or number of nearest neighbors, can be examined using the backscattering amplitudes of Mn, Ga, As and Be created using FEFF 8.2. The coordination number of the central Mn atom is then allowed to vary. This allows IFEFFIT to decrease the backscattering from the nearest neighbors to match the decrease in first shell magnitude. This led to unphysically low coordination numbers, indicating that this possibility is not the cause of the change in magnitude.

The second possibility can be ruled out through examination of the $k\chi$ data in Fig. 3. If the Debye-Waller factor was the cause of the decrease in the magnitude of the first shell then the expectation would be that the $k\chi$ -data would match for the first several oscillations, decrease at higher k values, and that the decrease would be more rapid for the materials with higher disorder. The samples with the largest decrease in first shell magnitude would have the most rapid decrease in amplitude of the $k\chi$ -data. However, this does not happen, which removes this possibility.

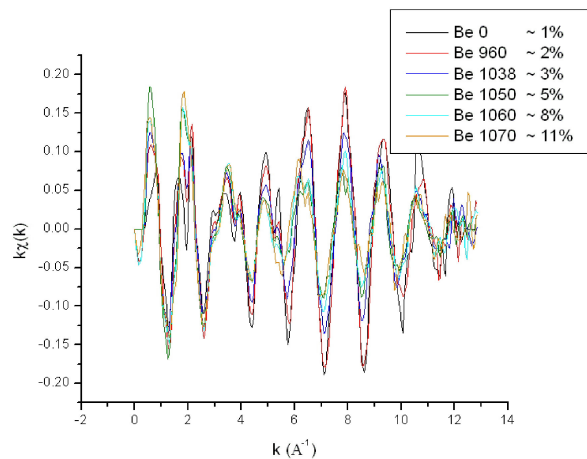


Fig. 3

The remaining possibility is then that the cause of the change in first shell magnitude is due to interference between the signals from different sites that the Mn ions occupy in the $\text{Ga}_{1-x-y}\text{Mn}_x\text{Be}_y\text{As}$. Attempts were made to fit the Be co-doped materials using a linear combination of the models mentioned earlier. These attempts were followed by creation of three part models based upon the models listed before. These three part models were successful in fitting the $\text{Ga}_{1-x-y}\text{Mn}_x\text{Be}_y\text{As}$ materials with higher concentrations of Be. However, for sample Be 0 which had no Be and Be 960 which had only one percent Be, single structure models did work better than any of the two or three part models. The best fits to the data were created using the Mn_{Ga} , $\text{Mn}_{\text{I-Ga}}$, and MnAs models.

Discussion

XAFS examinations of the Mn local structure in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ and $\text{Ga}_{1-x-y}\text{Mn}_x\text{Be}_y\text{As}$ materials have been conducted. The $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ materials, in which several Mn concentrations were examined, exhibited a structure of Mn substituting onto the Ga site within the GaAs lattice. All concentrations of Mn were able to be fitted with this substitutional model using up to three single scattering paths. The XAFS of the $\text{Ga}_{1-x-y}\text{Mn}_x\text{Be}_y\text{As}$ materials indicated the presence of multiple local structures for the Mn within the materials. The samples with small amounts of Be; Be 0, and Be 960 were found to be best modeled by the substitutional model using up to 3 shells. However, the samples with higher levels of Be; Be 1038, Be 1050, Be 1060, and Be 1070, required combinations of local structures. The percentage of Mn in the Mn_{Ga} local environment declined as a function of Be concentration, while the percentage of Mn in the $\text{Mn}_{\text{I-Ga}}$ and MnAs environments increased as a function of Be concentration. Additionally, there was no measurable sign of Mn-Mn or Mn-Be pairing.

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References

- [1] H. Ohno Science. **281**, 951 (1998).
- [2] H. Ohno J. Mag. Mat. **200**, 110-129 (1999).
- [3] K.M. Yu, W. Walukiewicz, T. Wojtowicz, I. Kuryliszyn, X. Liu, Y. Sasaki, and J.K. Furdyna, Phys. Rev. B **65**, 201303R (2002).
- [4] K.M. Yu, W. Walukiewicz, T. Wojtowicz, L.W. Lim, X. Liu, Y. Sasaki, M. Dobrowolska, and J.K. Furdyna, Appl. Phys. Lett. **81**, 844-846 (2002).
- [5] M. Newville, J. Syn. Rad. **8**, 322-324 (2001).
- [6] A.L. Ankudinov, B. Ravel, J.J. Rehr, and S.D. Conradson, Phys. Rev. B, **58**, 7565 (1998).
- [7] A.L. Ankudinov, C. Bouldin, J.J. Rehr, J. Sims, H. Hung, Phys. Rev. B, **65**, 104107 (2002).