Site Occupation Preferences in CdMnTeSe Quaternary Alloys. EXAFS Data Analysis

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Abstract. Site occupation preferences in zinc-blende $Cd_{1-x}Mn_xTe_{1-y}Se_y$ ($0.0 \leq x \leq 0.15$; $0.0 \leq y \leq 0.20$) quaternary alloys have been determined by the Extended X-ray Absorption Fine Structure (EXAFS) analysis. The experimental data have been collected with the use of synchrotron radiation for Se- and Mn-K edges. For both K edges, the preferences of manganese distribution around selenium have been observed and analyzed. The Mn-Se with Cd-Se, and Mn-Se with Mn-Te distances have been found as linearly dependent on concentration of Se and Mn. The average coordination numbers of Mn around Se versus Mn composition was approximated by the quadratic behavior instead of the linear dependence expected for the virtual crystal approximation, in which a full random distribution of elements in the sublattices is assumed. The obtained results support the preferential occurring of Mn and Se ions in the considered compounds. A structural and magnetic interaction reasons of the preferences were discussed.

Extended X-ray Absorption Fine Structure (EXAFS) analysis of the random ion distribution has been dated since the random cluster model (RCM) [1, 2] was applied to CdMnTe [1], $TeCdTe$, GaInSb and HgTeSe [3]. This model has been developed finally as a random iterative cluster model (RICM) by Wiedmann *et al.* [4] and applied to the analysis of the EXAFS data for $Zn_{1-x}Mn_xSe$ [5]. In quaternary alloys site correlations are expected to be larger than for ternaries because nearest-neighbor preferences are possible. They were observed in InGaSbAs compounds by Islam and Bunker [6]. They found clear preference of Ga-As pairs over In-As pairs.

The aim of this paper is to analyze discrepancies in random distribution of quaternary $Cd_{1-x}Mn_xTe_{1-y}Se_y$ diluted magnetic semiconductors. Experimental X-ray absorption measurements have been carried out with use of synchrotron radiation at the Adone Wiggler facility in Frascati utilizing the Si(111) and Si(220) channel-cut crystal monochromators [7]. The original samples were high purity monocrystalline binary CdTe, CdSe, MnTe and MnSe standard compounds, ternary CdMnTe and $Cd_{0.75}Mn_{0.25}Se$ and $Cd_{1-x}Mn_xTe_{1-y}Se_y$ ($x = 0.00, 0.05, 0.10, 0.15$; $y = 0.00, 0.10, 0.20$) quaternary compounds produced by Bridgman method. The binary compounds crystallize in three different structures; i.e. CdTe crystallize in zinc-blende structure, MnTe in hexagonal NiAs structure, CdSe in wurtzite structure and MnSe in NaCl structure. Quaternary compounds CdMnTeSe have well defined monophasic zinc blende structure only for low Mn and Se concentration. The phase diagram of this alloy system [8] shows wide two and three phase region in the center with narrow monostructure islands in the corners of phase diagram. Especially, the monophasic zinc-blende region extends for $x < 0.2$ when $y < 0.2$ and $x < 0.7$ when $y < 0.05$. All compositions of studied samples derived from this monophasic region. XAS measurements have been carried out on the Mn and Se K edges. The contribution of each edge to the absorption coefficient has been isolated by extrapolating the pre-edge region to higher energies by a Victoreen-like fit and by subtracting the fitted curves from the remaining experimental spectrum [9]. EXAFS data analysis for Mn and Se K edges was done applying the well known data reduction procedure described elsewhere [10]. In this work we are mainly interested in neighbor distribution in the first shell. The first shell data was filtered from fine structure using Fourier and back Fourier transform. The one shell parameters (N , R and σ) was obtained by non-linear least-squares fitting. Figure 1 shows the Mn-Se coordination as a function of Mn composition

Figure 1: Se neighborhood vs. Mn concentration

taken from *Se* *K*-edge. The solid line (square function) was fitted to experimental points for 5%, 10%, 15% of *Mn* with 10% of *Se* as well as to two border points for ternaries $CdTe_{0.9}Se_{0.1}$ and hypothetical zinc-blende $MnTe_{0.9}Se_{0.1}$ compounds. The consistent deviation from random distribution represented by dashed line is visible. This results are confirmed also by data obtained from *Mn* *K*-edge. This nonlinear dependence is analytically described by quadratic function. Taking into consideration the random distribution background expressed by $4x$ (dashed line on fig.1) the preference of the site occupation could be approximate as proportional to $x(1-x)$. This result is in very good agreement with Perkowitz *et al.* [11] expectations for the analytical dependence of the clustering parameter versus composition. For both *Se* and *Mn* *K*-edges the preference of *Mn-Se* pairing over *Cd-Se* and *Mn-Te* pairs was observed, respectively. The preference of the distribution of the Mn ion around Se is in agreement with tendencies anticipated from the lattice constants of the zinc blende binary compounds which fulfill inequality $CdTe > MnTe > CdSe > MnSe$. Also a local ordering in this alloy would be the strain energy which depends on the size of constituent atoms.

In a previous paper [12] we have generalized the model of Czyżyk *et al.* [2] to the case of quaternary alloys. The model is based entirely on the elastic bond-stretching and bond-bending forces and gives a satisfactory account for nearest-neighbor distances in these alloys. On the other hand, this very simple model does not describe properly the possibility of preferences.

Summarizing, we conclude that apart from the evident structural reason of the preferences which should be discussed in more advanced form, in the case of $CdMnTeSe$ the magnetic superexchange interaction which is larger in $CdMnSe$ than in $CdMnTe$ should be also considered [13].

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