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BAND STRUCTURE AND MAGNETIC PROPERTIES OF DO$_3$-TYPE $Fe_{3-x}V_xAl$ ALLOYS. SUPER-CELL APPROACH

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The electronic structure of $Fe_{3-x}V_xAl$ alloys was calculated using the super-cell methodology of alloy modeling. The concentration range of $x = 0.0 \div 1.0$ was investigated. For a concentration of $x = 0.0625$ the energy based analysis reveals that vanadium prefers to replace the Fe atom at sites with the octahedral coordination. It was found that the iron atoms coordinated by the eight nearest-neighbor Fe atoms preserve their high magnetic moment up to a concentration of $x = 0.9375$ even though the average total magnetic moment goes to zero. The relatively high ($\simeq -1.0\mu_B$) negative magnetic moment of V remains constant up to $x \simeq 0.5$. In the concentration range of $x = 0.75 \div 0.9375$ the gap at ε_F of the minority density of states is observed while the majority density of states displays a sharp peak structure at the Fermi energy. This feature suggests the heavy-fermion behavior of the Fe_2VAl compound.

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Because of unusual magnetic and electronic behavior the Heusler-type $Fe_{3-x}V_xAl$ alloys are the subject of the intensive experimental [1–3] and theoretical [4–7] study. The magnetic measurements have shown that for low vanadium concentrations ($x \lesssim 0.2$) the alloys are ferromagnets with the magnetic moment decreasing more sharply than the simple dilution related to the replacement of Fe moments with non-magnetic V atoms [1, 3]. The estimations based on the local environmental model [1] also result in the linear decrement of magnetic moment but the slope differs from the measured one. For the concentration range $0.3 \leq x \leq 0.6$ the magnetic moment decreases rapidly and for $x \geq 0.5$ the magnetostatic measurements [1, 2] do not show the long range magnetic order while the Mössbauer effect spectrum for $x = 0.6$ shows the presence of the Fe in the high-magnetic state [1]. The effect was attributed to the coexistence of the ferromagnetic and super-paramagnetic phases [1]. The anomalous semiconductor-like behavior of the electrical resistivity of Fe_2VAl was explained by the presence of the narrow energy pseudo-gap at the Fermi level, confirmed by the XPS measurements of the valence band [2]. Despite the presence of the energy gap the Fe_2VAl compound shows a large electronic specific-heat coefficient $\gamma = 14$ mJ mol^{-1} K^{-1} [3] related to the large mass enhancement; the effect is typical of the heavy-fermion systems [8]. The electronic structure of the Fe_2VAl compound and disordered $Fe_{3-x}V_xAl$ alloys have been the subject of the theoretical study [4–7]. The band structure calculations confirm the existence of the gap in the density of states of Fe_2VAl at the Fermi level but no explanation was found for the mass enhancement. It was suggested [4, 5] that the mass enhancement was due to the spin-fluctuations effect. The super-cell [5] and the KKR-CPA [7] calculations of the ordered and disordered $Fe_{3-x}V_xAl$ alloys showed that the iron atoms at sites coordinated by the eight nearest-neighbor Fe atoms had a strong, localized magnetic moment of magnitude in the range of ~ 2.2–$3.2\mu_B$.

The aim of the presented calculations is to extend the super-cell calculations reported in [5] to cover a wider vanadium concentration range in $Fe_{3-x}V_xAl$ and to analyze the dependence of the magnetic structure and the hyperfine parameters on the vanadium contents.

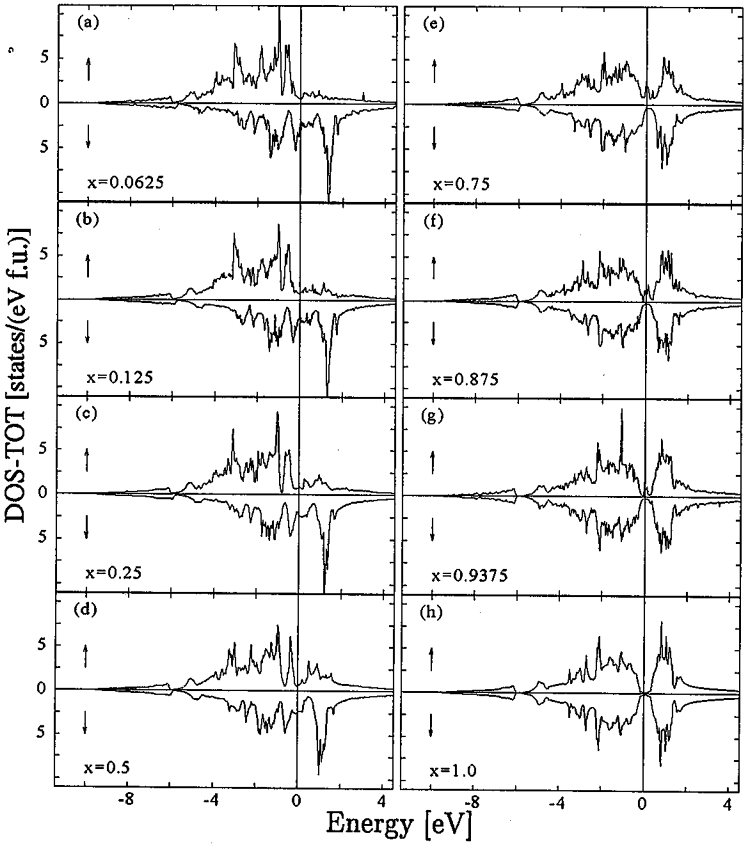

Fig. 1. The spin-resolved total density of states (DOS) of $Fe_{3-x}V_xAl$ for vanadium concentrations $x = 0.0625 \div 1.0$. The vertical solid line across all figures shows the Fermi energy.

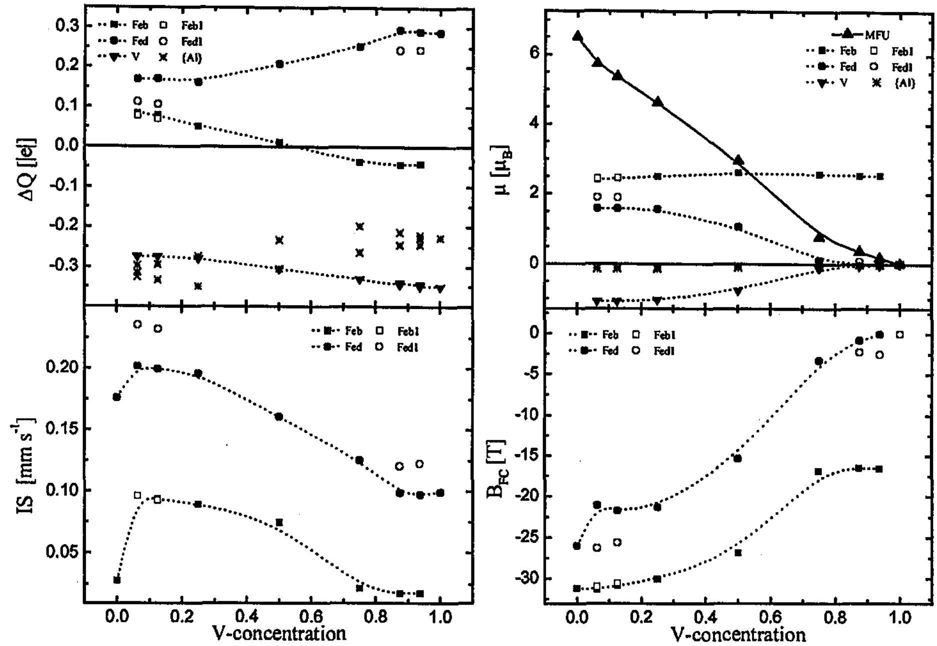

Fig. 2. Site resolved interatomic electron transfer (ΔQ), isomer shift (IS), partial (atomic) and total (per f.u.) magnetic moments, and Fermi contact hyperfine field (B_{FC}) versus vanadium concentration (negative values of ΔQ mean the transfer outward an atomic sphere).

In the DO$_3$-type structure iron atoms occupy three positions grouped into two different classes. The first class with one atomic position is occupied by Fe atom (denoted as Feb) coordinated by eight Fe atoms (octahedral coordination). The second class consists of two atomic positions is occupied by Fe atoms (called as Fed) surrounded by four Al and four Fe atoms (tetrahedral coordination). In the Heusler-type Fe$_2$VAl compound the Feb is replaced by the V atom. In order to investigate the non-stoichiometric compositions of Fe$_{3-x}$V$_x$Al the super-cells were constructed consisting of 2, 4, 8, and 16 underlying DO$_3$-type unit cells. The calculations were performed for V concentrations in the range of $x = 0.0625$–1.0. For some concentrations the disorder produced by the partial replacement of Feb atoms with V causes the splitting of the crystallographically equivalent positions into different crystallographic classes.

In the calculations the tight-binding linear muffin-tin orbital (TB-LMTO) method [9] in the atomic sphere approximation (ASA) was used. The correction for the Wigner–Seitz sphere overlap was taken into account. The exchange-correlation with gradient (non-local) corrections [10, 11] was utilized. The approach was scalar-relativistic with spin–orbit interaction to be neglected. The lattice parameters were taken from experiment [2] and the space-filling equal-radius atomic spheres were assumed [7]. The Fermi contact hyperfine field was calculated applying the relativistic formula given by Blügel et al. [12].

Figures 1 and 2 summarize the results of the calculations. The analysis of the partial (atomic and spin-resolved) densities of states showed that up to the concentration of $x = 0.5$ the DOS of $Fe_{3-x}V_xAl$ at the Fermi level (ε_F) is dominated by the minority spin d-states of Fed atoms. The contribution of the states from Feb and V atoms near the Fermi level is relatively small. For concentrations of $x = 0.75 \div 0.9375$ the Fed-d minority DOS contracts and shifts below ε_F. In the same time the strong hybridization of the majority-spin d-like states of Feb atoms with these of the nearest-neighbor Fed atoms gives rise to a sharp, relatively high peak of majority DOS at the Fermi level. This peak disappears for the stoichiometric Fe_2VAl compound and the pseudo-gap of ~ 0.7 eV wide develops at ε_F. This feature of the DOS in the vicinity of ε_F may suggest the heavy-fermion-type behavior of $Fe_{3-x}V_xAl$ Heusler-type alloys near the stoichiometry. The results presented in Fig. 2 can be summarized as follows: (1) A remarkable charge transfer of electrons from V and Al atoms to the Fed-group of atoms is observed which may explain the filling and shifting of the minority d-states of Fed atoms below ε_F; (2) The positive values of isomer shift on Fe atoms suggests the decrease in the s-state charge density at Fe sites; (3) The non-applicability of simple dilution law and the local environmental model in the explanation of magnetic moment variation in $Fe_{3-x}V_xAl$ for $x \leq 0.5$ is caused by the presence of the large, negative magnetic moment of V atoms; (4) The local magnetic moment of Feb atoms remains high for $x \geq 0.8$ even though the average magnetic moment of alloys almost disappears; (5) The non-zero hyperfine field observed experimentally for high concentrations of V can originate from the high-magnetic Feb atoms.

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