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## MAGNETOELASTIC PROPERTIES OF $\text{MnRhAs}_{1-x}\text{P}_x$ AND BAND STRUCTURE RESULTS FROM KKR-CPA

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The magnetoelastic phase transitions were observed in  $\text{MnRhAs}_{1-x}\text{P}_x$  system (space group  $P62m$ ) by X-ray measurements ( $80 \text{ K} < T < 300 \text{ K}$ ). For  $x = 0.33$  and  $x = 0.4$  contents an abrupt change of both lattice parameters and of the elementary cell volume at the metamagnetic transition was detected. The Korringa-Kohn-Rostoker electronic structure calculations performed for MnRhP compound show significant variations of densities of states at the Fermi level, while changing lattice data. The Korringa-Kohn-Rostoker-coherent potential approximation studies of disordered  $\text{MnRhAs}_{1-x}\text{P}_x$  system ( $x = 0.6, 0.8, 1.0$ ) were done at the ferromagnetic region. From our calculations we observe moving of bands near  $E_F$ , while substituting with isoelectronic metalloid, likely as while changing the crystal data in MnRhP.

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### 1. Introduction

The MnRhAs compound exhibits the antiferromagnetic ordering (AF) at low temperature range while the complex magnetic structure, where both anti-ferromagnetic and ferromagnetic (AF+F) phases coexist, is observed for higher temperatures [1, 2]. With alloying in disordered  $\text{MnRhAs}_{1-x}\text{P}_x$  system the hexagonal crystal structure in the whole range of concentration is maintained. The end-compound MnRhP, when replacing As by isoelectronic P, exhibits in contrast to MnRhAs, ferromagnetic (FM) ordering (up to  $T_c = 400 \text{ K}$ ). For the

intermediate concentration range of  $\text{MnRhAs}_{1-x}\text{P}_x$  system (at low temperature) the pure AF state is observed only for  $x \leq 0.5$ , when the ferromagnetic component appears, reaching FM ordering for  $x \geq 0.6$  (up to  $T_c$ ). The magnetic properties in  $\text{MnRhAs}_{1-x}\text{P}_x$  alloys are mainly due to the magnetic moment observed on the manganese (3*g*)-site ( $\approx 3\mu_B$ ). Nevertheless a small magnetic moment on Rh(3*f*) atoms ( $0.2 \div 0.3\mu_B$ ) was detected in parent compounds (at higher temperature range) [2, 3], which is not supported by our ground state Korringa-Kohn-Rostoker-coherent potential approximation (KKR-CPA) calculations. The metamagnetic phase transitions in  $\text{MnRhAs}_{1-x}\text{P}_x$  system can be induced by magnetic field or external pressure [4-6]. A similar effect (the preference of the FM ordering) was detected with the increase in the external pressure or the increase in the phosphorus content.

## 2. Experimental results

The magnetoelastic phase transition between the AF and the (AF+F) phases for the  $\text{MnRhAs}$  compound, confirmed by neutron diffraction measurements, is accompanied by the jump of both lattice constants ( $a$  and  $c$ ) and the elementary cell volume [1, 2]. The analysis of magnetoelastic effect was performed for  $\text{MnRhAs}_{0.67}\text{P}_{0.33}$  and  $\text{MnRhAs}_{0.6}\text{P}_{0.4}$  samples (near AF+F phase limit) with the use of the X-ray diffraction measurements at various temperatures. The temperature dependences of  $a$  and  $c$  lattice parameters were deduced from the X-ray diffraction patterns collected in the 90-300 K temperature range. The Rietveld technique was used for the refinement of the lattice parameters. Moreover, the  $x(3g)$  and  $x(3f)$  crystal structure parameters, locating Mn and Rh atoms in the unit cell, were determined both in the AF and the (AF+F) phases. No deviation from stoichiometry, within the experimental error  $\approx 1\%$  was found for the investigated samples. The values of  $a$  and  $c$  lattice constants, measured at room temperature, remain in good agreement with data previously found by Chaudouet [1]. The jump of the lattice parameters associated with the metamagnetic phase transitions were found for  $\text{MnRhAs}_{0.67}\text{P}_{0.33}$  and  $\text{MnRhAs}_{0.6}\text{P}_{0.4}$  systems. The values of  $\Delta a$  and  $\Delta c$  are of order 0.1% and 0.3%, respectively. The jump of the elementary cell volume was also established with  $\Delta V$  equals 0.4% and 0.2% for  $x = 0.33$  and  $x = 0.4$  samples, respectively (Fig. 1).

## 3. Band structure calculations

The electronic structure studies of  $\text{MnRhP}$  compound and  $\text{MnRhAs}_{1-x}\text{P}_x$  ( $x = 0.6, 0.8, 1.0$ ) were motivated by supposing a related variations of bands near the Fermi level with changing of the unit cell volume or alloy composition changes. The band structure calculations of the ordered systems were performed by the charge and spin selfconsistent Korringa-Kohn-Rostoker (KKR) method. The computations of  $\text{MnRhP}$  were carried out for the different experimental crystal data, inspired by pressure dependent phase diagram. The electronic structure of disordered  $\text{MnRhAs}_{1-x}\text{P}_x$  alloy using experimental crystal data, was calculated by KKR method with the coherent potential approximation (CPA) [7, 8]. The dispersion curves  $E(\mathbf{k})$  along the high symmetry direction in Brillouin zone (BZ) are

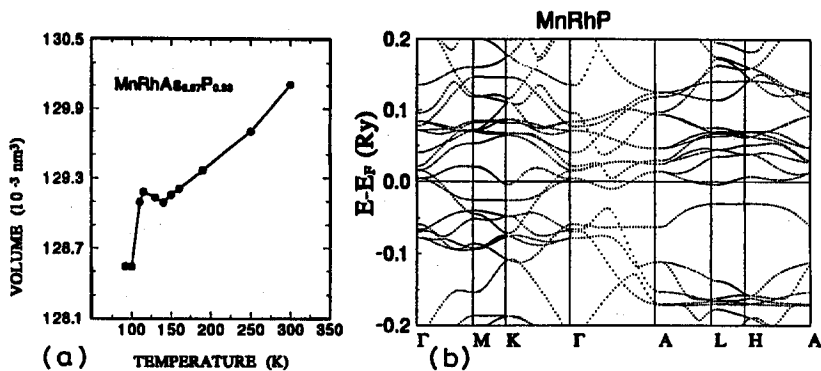


Fig. 1. (a) The unit cell volume variations versus temperature for  $\text{MnRhAs}_{0.67}\text{P}_{0.33}$ , (b) the spin-down dispersion curves  $E(\mathbf{k})$  near the Fermi level along high symmetry directions in BZ of  $\text{MnRhP}$  ( $a = 6.2446 \text{ \AA}$ ,  $c = 3.5943 \text{ \AA}$ ,  $x(3f) = 0.2617$ ,  $x(3g) = 0.5967$ ).

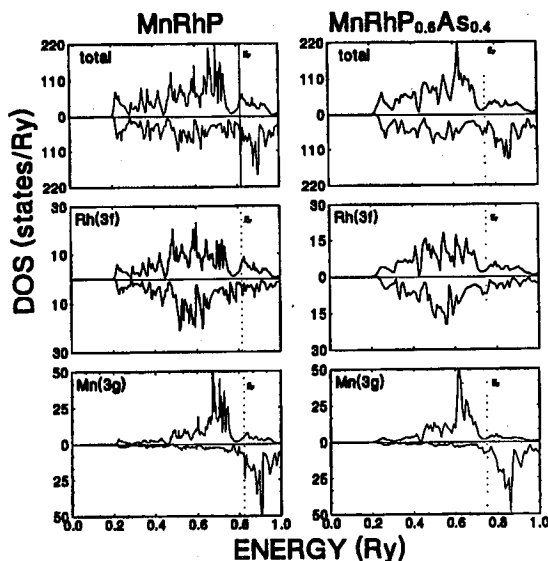


Fig. 2. The spin-projected total and component DOS for  $\text{MnRhP}$  compound and disordered  $\text{MnRhAs}_{0.4}\text{P}_{0.6}$  alloy ( $a = 6.351 \text{ \AA}$ ,  $c = 3.607 \text{ \AA}$ ).

also presented for three ordered compounds:  $\text{MnRhP}$ ,  $\text{MnRhP-mod}$  (with modified crystal data observed for  $\text{MnRhAs}_{0.4}\text{P}_{0.6}$ ) and  $\text{Mn}_3\text{Rh}_3\text{AsP}_2$ , which is close to the disordered  $\text{MnRhAs}_{0.33}\text{P}_{0.67}$  alloy. In comparison with  $\text{MnRhP}$ , where  $E_F$  lies at the sharp spin-down peak (bands are “wrapped” around  $E_F$ , Fig. 1), in  $\text{MnRhP-mod}$  as well as in the hypothetical  $\text{Mn}_3\text{Rh}_3\text{P}_2\text{As}$ , these bands are found above the Fermi energy. At the same time, the spin-up bands do not change significantly, which affects the increase in the total magnetic moment per Wigner-Seitz cell from  $9.02\mu_B$  in  $\text{MnRhP}$  to  $9.94\mu_B$  in disordered  $\text{MnRhAs}_{0.4}\text{P}_{0.6}$  and  $9.89\mu_B$

in MnRhP-mod, since the magnetic moment on Mn(3g) slightly increases from  $3.07\mu_B$  to  $3.38\mu_B$  and  $3.31\mu_B$ , respectively. At present, the neutron diffraction results are available only for the end-compounds. The measured atomic magnetic moment on Mn(3g) is  $3.1\mu_B$  in MnRhP and  $3.5 \div 3.6\mu_B$  in MnRhAs (note, this system does not show the FM structure). These values well correspond to the magnetic moments found by the KKR method,  $3.07\mu_B$  and  $3.42\mu_B$  (the band calculations performed for the single-cell MnRhAs system), respectively. The increase in the magnetic moment on Mn(3g), when passing from MnRhP to MnRhAs, is associated to the crossing of the spin-down subband above  $E_F$ . Observing the total and component DOS (Fig. 2), we see the decreasing of density of states at  $E_F$  with lowering of P content. This trend may be responsible for the preference of AF phase in samples with majority of As atoms, reaching the limit, when the Stoner criterion for the FM ordering is not maintained. From the KKR-CPA results we find the similar mechanism driving the electronic structure variations in disordered MnRhAs $_{1-x}$ P $_x$  system with concentration changes, as with changing the elementary unit volume in ordered MnRhP. In that sense the effect of moving bands near  $E_F$ , while substituting with isoelectronic metalloid in MnRhAs $_{1-x}$ P $_x$  system, may be interpreted in terms of "chemical pressure".

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