Magnetic Properties of Ta(Fe$_{1-x}$T$_x$)$_2$ with $T$=V, Cr, Mn, Co and Ni

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Abstract. The C14 Laves phase TaFe$_2$ is a paramagnet. The magnetic properties and phase diagrams of V, Cr, Mn, Co and Ni substituted systems for Fe in TaFe$_2$ have been investigated using magnetization and NMR measurements. Antiferro- or/and ferromagnetic states appeared by the V, Cr and Mn substitutions. In the Cr and Mn cases ferromagnetic states appeared first (very weak in Cr case) and then the both change to be antiferromagnetic. In the V case only antiferromagnetic state appeared from the beginning of substitution. The susceptibilities of the slightly V-substituted compounds were, however, exchange-enhanced at low temperatures. The above mentioned magnetic changes are qualitatively the same to the previously reported Nb(Fe$_{1-x}$T$_x$)$_2$ systems. The electronic condition of the appearance of ferro- and antiferromagnetism can systematically be interpreted in both systems.

1. Introduction

We have previously reported the magnetic states and phase diagrams of the Fe substituted C14 Nb(Fe$_{1-x}$T$_x$)$_2$ systems with 3d-elements, $T$=V, Cr, Mn, Co and Ni[1]. The NbFe$_2$ is a weak antiferromagnet below 10 K[2]. The antiferromagnetically ordered state is very delicate and apparently impaired by magnetic fields larger than 6 kOe or by a slight deviation of the composition from the stoichiometry[2]. The deviation towards both the Fe- and Nb-excess sides leads to a ferromagnetic order, suggesting the coexistence of ferro- and antiferromagnetic spin fluctuations. The antiferromagnetism in NbFe$_2$ collapses by the substitution of the 3d-element less than 1 at. % in all the cases. The Co and Ni substitutions make NbFe$_2$ less magnetic, while a weakly ferromagnetic state appears in steady at the low concentration region by the V, Cr and Mn substitutions. The ferromagnetism is weak in the V case ($T_C$≈26), medium in the Cr case ($T_C$≈34 K) and strong in the Mn case ($T_C$≈63 K). From the plots of Curie temperatures against the unite cell volume or the 3d-electron number in a molecule, the ferromagnetism of these systems have been found to be enhanced all at around common values, 161 Å (about 0.5 % increase against that of NbFe$_2$) or 11.75 of the number (0.25 decrease from that in NbFe$_2$), respectively[1]. The magnetic states of NbFe$_2$ and the substituted systems above mentioned, can be well explained by the theoretical investigations[3, 4].

Though the C14 TaFe$_2$ is paramagnetic[5], this compound is expected to be magnetically in the similar situation to NbFe$_2$, because tantalum locates just below niobium in the periodic table. The magnetic susceptibility of TaFe$_2$ increases at low temperatures, namely it is exchange-enhanced. The concentration deviation from the stoichiometry towards both Fe- and Ta-excess sides induces
ferromagnetism. It was reported that aluminium substituted Ta(Fe1-xAl)x2 system showed antiferromagnetism at x=0.05~0.2[6]. We have investigated also magnetic properties of Ta(Fe1-xTx)x2 with T=V, Cr, Mn, Co and Ni, using magnetization and NMR measurements.

2. Experimental procedure

Samples with x≤0.3 for vanadium, x≤0.35 for chromium, x≤0.4 for manganese, x≤0.3 for cobalt and nickel were prepared by arc melting in argon atmosphere and annealed at 1050 °C for four weeks. No other phase than the C14 was detected by X-ray diffraction. The lattice constants, a and c, of the V, Cr and Mn (x≤0.25) substituted compounds increase almost linearly with x and the ascending order of their slopes is from Mn, Cr to V substituted cases. In the case of Mn substitution, a and c do not increase with x for 0.25<x≤0.4. Though other phases than the C14 were not detected in the X-ray diffraction patterns for 0.25<x≤0.4, the samples with x>0.25 should contain other phase(s) as describing in section 3. The a and c of the Co and Ni substituted compounds decrease by the substitution.

Magnetization measurements were carried out from 4.2 to 300 K up to 10.18 kOe. 51V and 59Co NMR spectra were obtained using a phase-coherent spin-echo spectrometer under the swept external field at 10.1 MHz at various temperatures.

3. Experimental Results

Figure 1 shows the temperature dependence of magnetization of some Ta(Fe1-xV)x2 samples under an external field of 10.18 kOe. The magnetization of TaFe2 shows an increase at low temperatures. By the substitution of vanadium for iron, the increase becomes large at first (x=0.025) and then small. Though the temperature change of Arrott plots of magnetization for x=0.025 suggests no existence of spontaneous magnetization down to 0 K, the sample is expected to be in a nearly ferromagnetic state. At x=0.08~0.25 where the increases of magnetization at low temperatures are small, a shoulder or swell appears at a temperature in the magnetization-vs-temperature curves, suggesting existence of an antiferromagnetic ordering.

All the 51V-NMR spectra of some samples for x≥0.08 show a peak almost at the zero-shift position and the peak position does not change by changing temperature. On the other hand, the line widths of the spectra for x=0.08~0.25 increase with decreasing temperature below the temperature at which the magnetization-vs-temperature curve shows a shoulder or swell. With increasing x, the line width at 4.2 K increases up to x=0.125 and then decreases. These results of NMR measurements clearly indicates that the shoulders and swells are corresponding to the Néel temperatures. For x=0.05 a shoulder could barely be seen at around 40 K in the magnetization-vs-temperature curve. That should also correspond to the Néel temperature, because the NMR spectra also shows an increase of the line width at low temperatures. In addition, a small negative shift of the peak position was seen in this spectrum, showing the nearly ferromagnetic state in this compound. The Néel temperatures were plotted against x in figure 2. The substitution of vanadium for iron in TaFe2 has been found to occur an appearance of an antiferromagnetic state at around x=0.125, through a nearly ferromagnetic state at around x=0.025.

By the substitution of chromium, the magnetization at low temperatures increases strongly at low chromium concentrations of x≤0.07. From the Arrott plots of the magnetization, samples with x=0.02, 0.03, 0.05 are found to have spontaneous magnetizations at low temperatures below 10 K. At x≥0.1, though the magnetization increases with decreasing temperature as well, the increase is not strong. At 0.1≤x≤0.25, a shoulder appears at a temperature in the magnetization-vs-temperature curves. Taking into account the above-mentioned vanadium substituted case, the shoulders should be corresponding to antiferromagnetic orders. In figure 3 Néel temperatures at x≥0.1 were plotted with Curie temperatures at low chromium concentrations which were determined by the Arrott plots.

In the manganese substitution, the ferromagnetic state becomes strong and expands its region abruptly. Antiferromagnetism appeared at x≤0.225. Figure 4 shows the magnetic phase diagram of Ta(Fe1-xMnx)x2 system. The Curie temperatures determined from the Arrott plots, and the Néel temperatures for x=0.225 and 0.25 from the shoulder in the magnetization-vs-temperature curves.
The magnetization data for $x>0.25$, in Mn substituted case where the lattice constants do not increase, as mentioned in section 2, showed coexistence of an antiferromagnetic component and (a) strongly ferromagnetic phase(s). The former probably comes from C14 phase with $x \approx 0.25$, and the latter from unknown phase(s).

**Figure 1.** Temperature dependence of magnetization of $\text{Ta(Fe}_{1-x}\text{V}_x\text{)}_2$ at 10.18 kOe.

**Figure 2.** Magnetic phase diagram of $\text{Ta(Fe}_{1-x}\text{V}_x\text{)}_2$

**Figure 3.** Magnetic phase diagram of $\text{Ta(Fe}_{1-x}\text{Cr}_x\text{)}_2$

**Figure 4.** Magnetic phase diagram of $\text{Ta(Fe}_{1-x}\text{Mn}_x\text{)}_2$
Cobalt and nickel substitutions have been found to make TaFe$_2$ less magnetic from the magnetization measurements: the magnetizations and their temperature dependence becomes to be small. This result was confirmed about some cobalt substituted samples by the $^{59}$Co-NMR measurements. The $^{59}$Co-NMR spectra showed a structure, presumably originated from the nuclear quadrupole interaction. The line shape hardly changes both by the cobalt-concentration and the temperature of measurement.

4. Discussion
From the present experiments Ta(Fe$_{1-x}$Ta$_x$)$_2$ has been found to become magnetic by the V, Cr and Mn substitutions, while less magnetic in the Co and Ni cases. This is the same to the case of the substitutions of NbFe$_2$[1]. The vanadium substitution makes TaFe$_2$ to be antiferromagnetic ($x=0.05$–0.25), through a nearly ferromagnetic state at the slightly substituted region at around $x=0.025$. This is similar to the Al substituted case[6], as mentioned in section 1. In the Cr substitution weakly ferromagnetic state appears at $x=0.02$–0.05 and then that changes antiferromagnetic ($x=0.1$–0.25). In the Mn case ferromagnetic state which is much stronger than that in the Cr case, appears at a wide concentration region of $x=0.05$–0.215 and then antiferromagnetism follows ($x=0.225$ and 0.25).

The magnetic states for the V, Cr and Mn substitutions are different for the Ta(Fe$_{1-x}$Ta$_x$)$_2$ case and for the Nb(Fe$_{1-x}$Ta$_x$)$_2$ case. Antiferromagnetic states appeared by all the three substitutions in Ta(Fe$_{1-x}$Ta$_x$)$_2$, while not in Nb(Fe$_{1-x}$Ta$_x$)$_2$.

As for ferromagnetism, the way of appearance is, however, seen to obey the same rule that it becomes stronger by the substitution, from vanadium, chromium to manganese, in order in both Ta(Fe$_{1-x}$Ta$_x$)$_2$ and Nb(Fe$_{1-x}$Ta$_x$)$_2$ cases, though the strength of ferromagnetism is considerably different between the two cases. For the Nb(Fe$_{1-x}$Ta$_x$)$_2$ case, as described in ref. 1, this could be interpreted very well, if we assumed that ferromagnetism was enhanced around a certain value of the lattice constants or/and a certain value of the d-electron number. The same assumption can also interpret qualitatively about ferromagnetism in the present Ta(Fe$_{1-x}$Ta$_x$)$_2$ case. This must be since the band structure of TaFe$_2$ and NbFe$_2$ is similar each other. In the present case, the assumption is, however, not done very well as well as the Nb(Fe$_{1-x}$Ta$_x$)$_2$ case. Though its origin may simply be attributable to the difference of detailed band structure between TaFe$_2$ and NbFe$_2$, the existence of strong antiferromagnetism, we think, may suppresses the appearance of ferromagnetism in the Ta(Fe$_{1-x}$Ta$_x$)$_2$ systems, taking into account the results of the substituted case of strongly antiferromagnetic TiFe$_2$ [7, 8].

Recently, in some of the Nb(Fe$_{1-x}$Ta$_x$)$_2$ systems we have found the existence of very weakly antiferromagnetic regions at the more substituted region after the ferromagnetic one, which is essentially the same to the Ta(Fe$_{1-x}$Ta$_x$)$_2$ cases. All results on Ta(Fe$_{1-x}$Ta$_x$)$_2$ and Nb(Fe$_{1-x}$Ta$_x$)$_2$ will be reported in full in forthcoming publications.

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