

## CURIE-TEMPERATURE "SLATER-PAULING CURVE"

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**Abstract.** – The systematic variation of the Curie-temperature "Slater-Pauling curve" has been explained for the first time on the basis of the finite-temperature theory of the local environment effect. The peculiarity of the Curie temperatures in Fe-V, Fe-Ni, and Ni-Mn alloys is elucidated by using the effective exchange couplings.

Magnetic properties of transition metal alloys have extensively been investigated over fifty years. Experimental data for the ground state magnetization curves have been summarized as the Slater-Pauling curves [1]. The systematics of these curves and the deviations from the curves have been explained by many theoretical investigators [2], though there are still some difficulties in the description of the Fe-Ni and Fe-Mn alloys. On the other hand there was no theoretical investigation for the systematic variations of the finite-temperature properties. This is because the simple Stoner model could not describe the localized features at finite temperatures in the transition metal alloys. Recent theoretical progress [3] however enabled us to investigate the finite-temperature magnetism in alloys. So far we have reported the results of the calculations for Fe-Ni [4], Ni-Mn [5], Fe-V [6], and Fe-Cr [7] alloys. In this presentation we report the results of the first systematic calculations for the Curie-temperature Slater-Pauling curves as well as the Slater-Pauling curves, including the new results for Fe-Co, Co-Ni, and Ni-Cu alloys.

We start from the degenerate-band Hubbard model, and adopt the finite-temperature theory of the local environment effect (LEE) developed by Kakehashi [4, 7]. The theory takes into account the fluctuations of the local moments with respect to the atomic configuration as well as the thermal spin fluctuations by making use of the method of the distribution function [8] and the static approximation to the functional integral method [9]. In particular it describes the itinerant-electron spin glass. The theory reduces essentially to the single-site spin fluctuation theory by Hubbard and Hasegawa in the pure-metal limit [9]. In alloys the theory selfconsistently takes into account a large number of atomic and magnetic configurations on the neighboring atoms (e.g. 16777216 for the fcc and 65536 for the bcc).

The input parameters are the d electron number, d-band width, the density of states in noninteracting systems, and the effective exchange parameter. The first three parameters are taken from the band calculations. The last one is chosen to reproduce the experimental ground-state magnetization. Calculated Curie

temperatures are then 1200 K for Fe, 2500 K for Co, and 1100 K for Ni.

Figure 1 shows the Slater-Pauling curve in both theory and experiment. Calculated curves in Fe-V, Fe-Cr, Fe-Co, bcc Fe-Ni, Co-Ni, and Ni-Cu approximately follow the straight lines with the slope 45° in agreement with the experimental data. A branch in Ni-Mn is due to the antiferromagnetic coupling between Mn local moments. In Fe-Ni alloys the magnetic coupling between the Fe local moments changes from the ferromagnetic to the antiferromagnetic with the decrease of their amplitudes. This causes a rapid deviation from the Slater-Pauling curve. In both cases the self-consistent determination of the local moments in various local environments is essential to obtain the continuous magnetization *vs.* concentration curves. The single-site theory with use of the coherent potential approximation leads to the first order transition to the paramagnetic state in Fe-Ni alloys in contradiction to the experimental data [10].

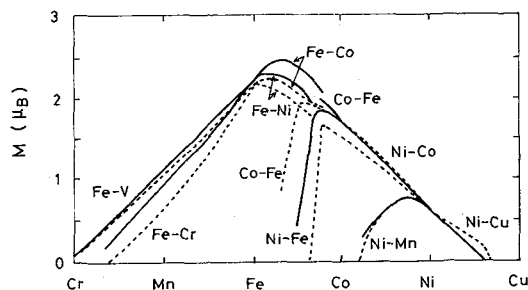


Fig. 1. – Theoretical (dashed curves) and experimental (solid curves) Slater-Pauling curves. The calculations are performed at 150 K.

In the hcp Fe-Co alloys a structural transition to the bcc takes place at 30 at. % Fe experimentally. Nevertheless we performed the calculations for the fcc Fe-Co alloys over all concentrations to understand the nature of the ferromagnetism in the close-packed systems. The result shows that the ferromagnetic instability occurs at 50 at. % Fe (see Fig. 1). This is caused by the same mechanism as in the Fe-Ni alloys.

The itinerant-electron spin-glass also appears after disappearance of the ferromagnetism. These theoretical predictions might be realized experimentally by stabilizing the closed packed alloys using the thin film technique or adding the third element into the alloys.

The calculated Curie-temperature Slater-Pauling curves are shown in figure 2 with the experimental data [11]. Although the present theory describes currently best the 3d transition metal alloys assuming phenomenological exchange energy parameters, it still overestimates the absolute values of  $T_C$  by a factor of 1.5-2.0 because of the neglect of the transverse degree of freedom of the local moments and the magnetic short range order [12]. The calculated curves are therefore renormalized by  $T_C$  in one of the constituent metals. Resulting curves explain well the experimental data.

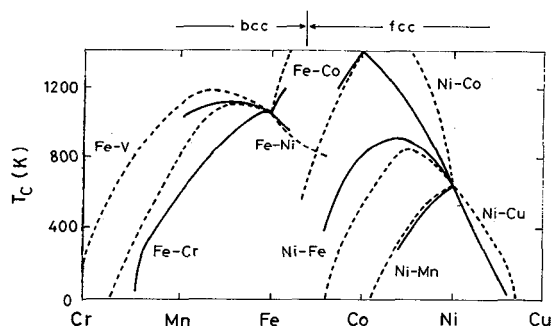


Fig. 2. — The Curie-temperature Slater-Pauling curves in the theory (dashed curves) and experiment (solid curves) [11].

Analytical expression of  $T_C$  has not yet been found. Thus it is not easy to argue in details the role of the electronic structures in  $T_C$ . A way to understand the  $T_C$  curves is to calculate the nearest-neighbor effective exchange coupling  $J_{\alpha\tau}$  between local moments on atoms  $\alpha$  and  $\tau$ . Since the Rhodes-Wohlfarth ratios of the alloys under consideration are rather close to one, the analyses based on such exchange couplings are expected to give a qualitative explanation to the  $T_C$  curves. In what follows we elucidate the concentration dependence from this viewpoint.

One of the important feature of itinerant magnetism is that the couplings  $\{J_{\alpha\tau}\}$  depend on the concentration. This explains the maximum of  $T_C$  in Fe-V alloys; it is caused by the enhancement of the coupling  $J_{FeFe}$  due to alloying with increasing V concentration. (Note that the concentration dependence was explained by the inequality  $|J_{FeV}| > |J_{FeFe}|$  independently of concentration in the localized model, while the present calculation shows that  $|J_{FeV}| \lesssim |J_{FeFe}|$ ). The same type of alloying effect is also seen in the bcc Fe-Ni

alloys, where  $J_{FeFe}$  rapidly decreases up to 10 at. % Ni with increasing Ni concentration. This effect decreases  $T_C$  in spite of the fact that  $J_{FeNi} > J_{FeFe}$ . On the other hand such alloying effect is no longer found in the bcc Fe-Co alloys;  $J_{CoCo} > J_{FeCo} > J_{FeFe} > 0$ , and  $J_{FeFe}$  gradually increases with increasing Co concentration. Thus  $T_C$  in Fe-Co alloys increase linearly with increasing average electron number. The maxima in the curves for fcc Fe-Ni and Fe-Co-Ni alloys are roughly explained by a rigid-band picture [13]. The Ni-Mn alloys show a simple decrease of  $T_C$  with increasing Mn concentration though the magnetization shows a maximum at 10 at. % Mn. This is because the former is determined by the inequality  $J_{NiNi} > J_{NiMn}$  while the latter results from the fact that the Mn local moments parallel to the magnetization are much larger than the Ni.

In conclusion we have explained qualitatively or semiquantitatively the Curie temperature Slater-Pauling curve as well as the Slater-Pauling curve on the same basis of the microscopic theory of the LEE. This is, we believe, a milestone in the field of magnetism in alloys. The different behavior between the Slater-Pauling curves and the Curie-temperature Slater-Pauling curves is explained by the alloying effects on the two kinds of degrees of freedom i.e. the local moments and the magnitude of the couplings between them. The systematic variation of the effective Bohr magneton numbers will be discussed elsewhere.

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